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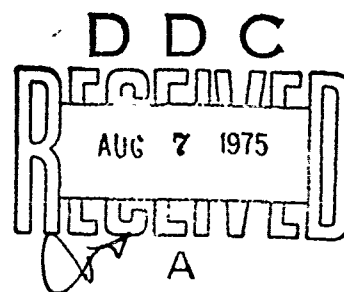
SHOCK-TUBE DIAGNOSTICS UTILIZING LASER RAMAN SPECTROSCOPY

BY  
A.A. Boiarski

17 APRIL 1975

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vibrational excitation times of oxygen in air, along with temperature and density measurements for oxygen and nitrogen in air were obtained over a range of postshock conditions. The measured densities and temperatures were found to be in good agreement with shock-wave theory. An excitation time measurement also agreed well with shock-tube data obtained with other diagnostic methods. In general, the results indicated that the Raman scattering technique was accurate at elevated temperatures and should prove useful in determining the thermochemical state of gases in continuous flows as well as in shock tubes and other impulse facilities.

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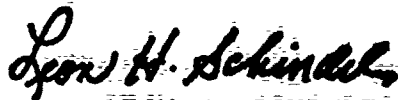
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## SHOCK-TUBE DIAGNOSTICS UTILIZING LASER RAMAN SPECTROSCOPY

This report documents measurements of high temperature gas properties behind a shock wave utilizing the laser Raman scattering, LRS, technique. The data was obtained at known postshock conditions in order to calibrate the experimental equipment for gasdynamic laser cavity diagnostics which were to follow. Since the IRS method is not as well known as other measurement techniques, a rather complete description of the required spectroscopic theory is also presented.

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LEON H. SCHINDEL  
By direction

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## SYMBOLS

$A(\Delta\lambda)$	apparatus function
$B_e$	rotational constant, $\text{cm}^{-1}$
$B_v$	rotational constant for vth vibrational state, $\text{cm}^{-1}$
$c$	velocity of light, $\text{cm/sec}$
$D_e$	rotational constant, $\text{cm}^{-1}$
$D_v$	rotational constant for vth vibrational state, $\text{cm}^{-1}$
$E$	energy, joules
$\Delta E$	quantized energy change, joules
$F(J)$	rotational term value, $\text{cm}^{-1}$
$G(v)$	vibrational term value, $\text{cm}^{-1}$
$g_J$	spin degeneracy factor
$h$	Plancks' constant, joule-sec
$I$	Raman intensity
$\bar{I}$	normalized Raman intensity
$I_B$	background light intensity
$I_L$	arbitrary value of incident laser intensity
$I_R$	arbitrary value of measured Raman intensity
$I_R(\lambda)$	computed Raman intensity
$J$	rotational quantum number
$\Delta J$	change in $J$ during Raman transition
$K$	cross section constant

$\ell$	scattering volume length, cm
$N_0$	incident photon flux, $\text{sec}^{-1}$
$N_{v,J}$	theoretical Raman line intensity, $\text{sec}^{-1}$
$N_{\lambda}^p$	computed Raman intensity flux, $\text{sec}^{-1}$
$n$	gas number density, $\text{cm}^{-3}$
$p$	gas pressure, torr or atm
$p_1$	preshock gas pressure, torr
$Q$	state sum
$S_J$	rotational line strength factor
$S_v$	vibrational band strength factor
$T$	temperature, $^{\circ}\text{K}$
$t$	time, sec
$v$	vibrational quantum number
$\Delta v$	change in $v$ during Raman transition
$W_s$	shock speed, meters/sec
$\alpha_e$	rotational constant, $\text{cm}^{-1}$
$\beta_e$	rotational constant, $\text{cm}^{-1}$
$\gamma$	ratio of specific heats
$\Delta$	wavelength correction factor, $\text{\AA}$
$\eta_i$	mole fraction of species $i$
$\lambda$	wavelength, $\text{\AA}$
$\nu_0$	incident laser frequency, $\text{cm}^{-1}$
$\nu$	frequency, $\text{cm}^{-1}$
$\Delta\nu_{v,J}$	frequency shift, $\text{cm}^{-1}$
$\rho$	gas density, $\text{grams/cm}^3$
$\sigma_{v,J}$	scattering cross section, $\text{cm}^2/\text{particle-sr}$

$\tau$	excitation time, sec
$\Omega$	solid angle of collection optics, steradians
$\omega_e$	vibrational constant, $\text{cm}^{-1}$
$\omega_e x_e$	vibrational constant, $\text{cm}^{-1}$
$\omega_e y_e$	vibrational constant, $\text{cm}^{-1}$
$\chi_{v,J}$	related to population density

Subscripts

A	actual value
C, Calib.	calibration condition
D	delay
F.L.	laser flash lamp
I	indicated value
J	rotational state
L	laser value
Lab	laboratory fixed coordinates
m	molecular property
max	maximum intensity
o	incident laser value
p	peak value
Q.S.	laser Q-switch
R	Raman value
S	scan value
rot, r	rotational energy mode
STD	standard condition
T	theoretical value
V-T	vibration-translation energy exchange process

vib, v	vibrational energy mode
w	wave fixed coordinates
1	preshock, also integer label
2	postshock, also integer label
4	driver gas property

## I. INTRODUCTION

The development of non-perturbing optical gas diagnostic techniques has received much attention in the past few years. This renewed interest is mainly due to the advent of modern high powered lasers and their associated electro-optical equipment. One such diagnostic method of current interest is the laser Raman scattering (LRS) technique.

Several experiments have been reported in which the LRS method was used to determine the thermochemical state of a test gas. The number density of various species was measured at low temperatures in both static and flowing gases.<sup>1-3</sup> High temperature Raman scattering measurements were also obtained for temperatures near 1700°K in H<sub>2</sub>/O<sub>2</sub>-air flames.<sup>4</sup> Several practical applications of LRS include the measurement of N<sub>2</sub> Raman scattering in an arc-heated wind tunnel environment<sup>5</sup> and pollutant density measurements in a gas turbine exhaust.<sup>6</sup>

---

<sup>1</sup>Hartley, D., "Transient Gas Concentration Measurements Utilizing Laser Raman Spectroscopy," AIAA Journal 10, pp. 687-689, May 1972

<sup>2</sup>Kellam, J. M. and Glick, M. M., "Gas Density Measurements in a Jet Using Raman Scattering," AIAA Journal 10, pp. 1389-1391, October 1972

<sup>3</sup>Widhopf, G. F. and Lederman, S., "Specie Concentration Measurements Utilizing Raman Scattering of a Laser Beam," AIAA Journal 9, pp. 309-316, February 1971

<sup>4</sup>Lapp, M., Goldman, L. M. and Penney, G. M., "Raman Scattering from Flames," Science 175, pp. 1112-1115, March 1972

<sup>5</sup>Boiarski, A. A. and Daum, F. L., "An Application of Laser-Raman-Spectroscopy to Thermochemical Measurements in an Arc Heated Wind Tunnel Flow," Aerospace Research Lab. Report ARL 72-0126, December 1972

<sup>6</sup>Bresowar, G. E. and Leonard, D. A., "Measurement of Gas Turbine Exhaust Pollutants by Raman Spectroscopy," AIAA/SAE 9th Propulsion Conference Paper No. 73-1276, Las Vegas, Nevada, 5-7 November 1973

The success of these experiments has prompted interest in the application of LRS as a diagnostic tool for shock tube research. White<sup>7</sup> suggested using Raman scattering as a possible means of making a noninterfering point measurement of vibrational and/or rotational temperatures behind an incident shock wave. He concluded that the species concentrations of individual components in a gas mixture could be measured. Also, these species measurements, as well as energy mode population measurements, could be made whether or not thermochemical equilibrium existed.

An attempt was made by Milling<sup>8</sup> to use Raman scattering as a diagnostic tool for measuring CO<sub>2</sub> dissociation rates in a shock tube experiment. No results were obtained, however, due to the fact that the CO<sub>2</sub> scattering cross section which he assumed was a factor of approximately ten times greater than a more recent measurement indicates.<sup>9</sup>

It was the objective of the present study to observe Raman scattering from high temperature O<sub>2</sub> and N<sub>2</sub> behind an incident shock wave in air. The postshock gas temperature and density were determined from the Raman technique and compared with known shock tube values over a relatively wide range of conditions. Besides this valuable high temperature calibration, the experiment also investigated the applicability of the LRS technique to shock tube excitation time measurements and general high temperature gas diagnostics.

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<sup>7</sup>White, D. R., "Shock Tube Studies of Nitrogen Vibrational Relaxation and Methane Oxidation," Aerospace Research Lab. Report ARL 70-0107, June 1970

<sup>8</sup>Milling, R. W., "Use of Time Resolved Raman Scattering for the Measurement of Dissociation Rates in a Shock Tube," Seiler Research Lab. Report SRL 69-005, June 1969

<sup>9</sup>Penney, C. M., Goldman, L. M. and Lapp, M., "Raman Scattering Cross Sections," Nature Physical Science 235, No. 58, pp. 110-112, 7 February 1972

## II. DETERMINATION OF THE THERMOCHEMICAL STATE OF A SHOCK HEATED GAS

### A. GENERAL DISCUSSION

When light passes through a gas, weak randomly directed radiation at the same frequency will appear due to scattering of the light by the gas molecules. The blue of the sky represents such an effect. In 1928 Raman<sup>10</sup> observed the spectrum of the light scattered from many different gases and liquids. Using a monochromatic source of frequency,  $\nu_0$ , he noted that the scattered light not only contained this incident frequency (i.e., Rayleigh scattering) but also several shifted frequencies,  $\nu_0 - \nu_1$ ,  $\nu_0 - \nu_2$ , and others,  $\nu_0 + \nu_1$  and  $\nu_0 + \nu_2$ . It was found that  $\nu_1$  and  $\nu_2$  were frequencies which were characteristic of the scattering molecule. That is, for a particular molecule, the shifted frequencies were different than frequencies obtained for any other molecule. This frequency shift phenomenon became known as the Raman effect. The radiation shifted toward lower frequencies is called "Stokes" radiation and the weaker radiation at higher frequencies, "anti-Stokes." Since its discovery, the Raman scattering effect has been examined for many different molecules and has been an important tool used by physicists for studying molecular structure at known densities and temperatures.

The Raman phenomena can be qualitatively explained by considering the molecule-photon interaction from the standpoint of conservation of energy. The incident photon at frequency  $\nu_0$  and energy  $E_0 = h\nu_0$  interacts with a molecule having energy  $E_m$ . This inelastic interaction produces a scattered photon of a different energy,  $E_{\text{Raman}} = h\nu_{\text{Raman}}$ , at a shifted frequency  $\nu_{\text{Raman}}$ . The energy differential is accounted for through a gain or loss of energy by the molecule. Since the molecular energy is quantized, only discrete amounts of energy may be exchanged in the interaction. Hence, the molecule must have an energy  $E_m'$  after the interaction which differs from its initial energy by a specified amount  $\Delta E_{\text{rot-vib}}$ . This energy differential is a known function of the bound energy states of the particular scattering molecule. Balancing energy before and after the interaction, the photon energies (i.e., frequencies) observed in Raman scattering must then correspond to the photon energy of the incident radiation shifted by an amount which is characteristic of the scattering molecule.

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<sup>10</sup>Raman, C. V., Indian Journal Physics 2, p. 287, 1928

The amount of frequency shift is given by:

$$\nu_0 \pm \nu_{\text{Raman}} = \frac{\Delta E_{\text{rot-vib}}}{hc} \quad (1)$$

At first there seems to be a myriad of possibilities for the scattered photon frequencies due to the many possible molecular vibrational energy states, denoted by quantum number  $v$ , each having many associated rotational energy levels, denoted by the quantum number  $J$ . However, there are selection rules which govern molecular energy transitions that can occur between various energy levels. The selection rules for a diatomic molecule undergoing Raman transitions are:

$$\Delta v = 0, \pm 1 \text{ (for vibrational transitions)}$$

and

(2)

$$\Delta J = 0, \pm 2 \text{ (for rotational transitions)}$$

These selection rules limit the number of allowed frequency shifts tremendously. However, in this study even fewer transitions will be investigated since only the  $\Delta v = +1$  vibrational and  $\Delta J = 0$  rotational transitions are important. These transitions give rise to the so-called Stokes vibration-rotation Q-branch Raman effect.

Even with the above restrictions, many transitions are still possible. This becomes evident, for example, when considering the Stokes Q-branch  $v=0$  to  $v=1$  vibrational transition, where  $J=1$  to  $J=1$ ,  $J=2$  to  $J=2$ , etc. rotational transitions can occur as well as the  $J=0$  to  $J=0$ . Due to small differences in rotational energy level spacing, the resulting frequencies are quite close but distinguishable. Also,  $v=1$  to  $v=2$ ,  $v=2$  to  $v=3$ , etc. vibrational Q-branch transitions can occur with all the  $\Delta J = 0$  transitions included in each  $\Delta v = 1$  vibrational transition. The various vibrational transitions are easily distinguished due to molecular anharmonicity. Hence, 1000-2000 Raman Q-branch transitions may be considered in the analysis of the Stokes rotation-vibration spectrum.

#### B. ROTATION-VIBRATION LINE INTENSITIES

Smekal<sup>11</sup> predicted the Raman effect in 1923 and since that time the theory of Raman scattering has become well developed from both

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<sup>11</sup>Smekal, A., "Zur Quantentheorie der Dispersion," Naturwiss 11, p. 873, September 1923



the quantum mechanical and the semi-classical viewpoints.<sup>12,13</sup> Hence, only the results of the detailed theoretical investigations will be presented in this report.

The observed Raman intensity flux in photons/second, resulting from a particular Stokes rotation-vibration transition is given by:

$$N_{v,J} = N_0 \chi_{v,J} \sigma_{v,J} \Omega \ell n \quad (4)$$

where  $N_0$  is the number of incident photons/second,  $\ell$  is the optically observed length of the laser beam in centimeters,  $\Omega$  is the solid angle of the collection optics in steradians,  $n$  is the number of molecules per cubic cm in the gas sample and  $\chi_{v,J}$  is related to the fraction of molecules in energy state  $E_{v,J}$ . Equation (4) shows that collected intensities vary directly with the incident photon flux. This fact explains why the advent of modern high intensity lasers has increased interest in Raman scattering as a practical diagnostic technique. Also, the Raman intensity is directly proportional to gas number density so measurements can be used to determine unknown density conditions.

Further examination of Equation (4) shows that the collected intensity is directly related to several experimental factors, namely, collector solid angle and observed laser beam length. Since the Raman photon flux is scattered nearly uniformly into  $4\pi$  steradians, a larger collector implies a greater collection efficiency. The scattering volume (i.e., volume from which collected photons originate) is defined by the laser beam cross section dimensions and the field of view of the optics. The volume varies linearly with the square of beam diameter and observed beam length. This is an important aspect of the Raman technique since it indicates that the measurement is truly a point determination of gas properties within the scattering volume. By focusing the laser beam and using optical stops to obtain a small observed length, the spatial resolution can be improved. However, Raman signal intensity is sacrificed when smaller and smaller field stops are employed.

Finally, the relation of the Raman scattering intensity to energy state population implies that detailed information can be obtained on population distributions in a gas. For non-Boltzmann conditions such as would exist in a laser cavity, a population distribution measurement is important. However, when Boltzmann statistics prevail, determination of the temperatures of various molecular modes is critical.

<sup>12</sup>Kramers, H. A. and Heisenberg, W., "Über die Streuung von Strahlung durch Atome," Z. Physik 31, p. 681, January 1925

<sup>13</sup>Placzek, G., "The Rayleigh and Raman Scattering," UCLR-Trans-526(L), translated from a publication of the Akademische Verlagsgesellschaft G.M.B.H., Leipzig, 1934

The symbol  $\sigma_{v,J}$  in Eq. (4) is the scattering cross section (i.e.,  $\text{cm}^2/\text{particle-sr}$ ) or transition probability for the particular  $v \rightarrow v+1$ ,  $J \rightarrow J$  transition and is written as:

$$\sigma_{v,J} = K (\nu_0 - \Delta\nu_{v,J})^4 g_J S_v \quad (5)$$

where  $K$  is a constant which can be theoretically estimated and experimentally measured. The frequency shift  $\Delta\nu_{v,J}$  of a Stokes Raman Q-branch line which appears in Eq. (5) is given by Reference 14 as:<sup>14</sup>

$$\Delta\nu_{v,J}^Q = \Delta\nu_v - \alpha_e J(J+1) - \beta_e J^2(J+1)^2 \quad (6)$$

where:

$$\Delta\nu_v = \omega_e - 2\omega_e x_e(v+1) + \omega_e y_e(3v^2 + 6v + 13/4) \quad (7)$$

In Equations (6) and (7) above,  $\omega_e$ ,  $\omega_e x_e$  and  $\omega_e y_e$  are vibrational constants and  $\alpha_e$ ,  $\beta_e$ , are rotational constants of the scattering molecule. The values of the above parameters used in the calculations reported in this text were taken from Reference 14. The symbol  $g_J$  in Eq. (5) is the spin degeneracy factor, which depends on the symmetry properties of a particular molecule and whether  $J$  is an even or odd integer.  $S_v$ , the vibrational band-strength factor, is given by:

$$S_v = v+1 \quad (8)$$

From Eq. (8), it can be seen that a  $v=1$  to  $v=2$  transition has twice the probability of occurring than the  $v=0$  to  $v=1$ . For a Boltzmann population distribution,  $\chi_{v,J}$  is a function of the vibrational temperature  $T_v$  and the rotational temperature  $T_r$  of the scattering molecule and is written:

$$\chi_{v,J} = \frac{S_J e^{-\frac{1.44F(J)}{T_r}} e^{-\frac{1.44G(v)}{T_v}}}{Q_{\text{rot}} Q_{\text{vib}}} \quad (9)$$

<sup>14</sup> Herzberg, G., Spectra of Diatomic Molecules, D. van Nostrand Company, Inc., New York, pp. 552-561, 1950

where the state sums  $Q_{\text{rot}}$  and  $Q_{\text{vib}}$  are given as:

$$Q_{\text{rot}} = \sum_J (2J+1) e^{-\frac{1.44F(J)}{T_r}} \quad (9)$$

$$Q_{\text{vib}} = \sum_v e^{-\frac{1.44G(v)}{T_v}}$$

The functions  $F(J)$  and  $G(v)$  which appear in Eq. (9) are so-called term values and are written as:

$$F(J) = B_v J(J+1) - D_v J^2 (J+1)^2 \quad (10)$$

and

$$G(v) = \omega_e (v+1/2) - \omega_e x_e (v+1/2)^2 + \omega_e y_e (v+1/2)^3$$

In Eq. (9)  $S_J$  is the line strength factor for Q-branch transitions and is given by:

$$S_J = (2J+1) \quad J=0,1,2,\dots \quad (11)$$

In Eq. (10)  $B_v$  and  $D_v$  are written as:

$$B_v = B_e - \alpha_e (v+1/2) \quad (12)$$

$$D_v = D_e + \beta_e (v+1/2)$$

Again  $B_e$  and  $D_e$  are rotational constants of the scattering molecule (Ref. 14).

The above equations show that the Raman line intensities are complex functions of number density, rotational temperature, vibrational temperature and various molecular constants and known experimental factors. Further, Raman frequency shifts are uniquely dependent on the scattering molecule. Hence, a measurement of the frequency

spectrum can yield an identification of the molecules which are present in the test sample. Also, the concentration of each constituent and the rotational and vibrational temperatures of each molecule species can be determined from a spectral intensity analysis.

### C. TEMPERATURE DETERMINATION

In the above equations the Raman spectrum (i.e.,  $N_{v,J}$  vs.  $\Delta v_{v,J}$ ) was shown to be a function of the rotational and vibrational population distributions of the molecules in a gas. For Boltzmann population conditions, the equations imply that a measurement of the spectrum would yield information concerning the rotational and vibrational temperatures independently. Hence, thermodynamic non-equilibrium conditions (i.e.,  $T_v \neq T_r$ ) could be investigated. The above statements come as no great surprise to one versed in other spectroscopic techniques, for the power of spectroscopy has been its ability to determine detailed thermochemical information about molecules in a test gas. The following describes how Raman spectroscopic information can be utilized in this respect.

From the above equations, the temperature dependence of the Q-branch Raman rotation-vibrational lines of molecular nitrogen was calculated for illustrative purposes. The results for the even rotational lines at  $T_v = T_r = 300^\circ\text{K}$  and  $1000^\circ\text{K}$  are plotted in Figure 1. A shift in the peak rotational intensity within the (0,1) rotation-vibration band at  $2300\text{ cm}^{-1}$  occurs as the temperature increases. This so-called "blue shift" is due to the increase in population of the upper rotational states with increasing rotation temperature. Also, at  $1000^\circ\text{K}$  a small set of frequency shifts appear at about  $2300\text{ cm}^{-1}$  resulting from the ( $v=1 \rightarrow v=2$ ) or (1,2) Q-branch transitions. These upper state Raman transitions appear in the spectrum due to the increase in the population of the upper vibrational states with increasing vibrational temperature.

In practice, a spectrum such as that shown in Figure 1 would be difficult to obtain due to the high resolution required to give individual rotational line intensities and due to the overlap of rotational lines of adjacent Q-branches. Hence, resolved spectra will not normally be obtained experimentally. To compute the unresolved spectrum, the Raman line intensities given above must be convoluted with the apparatus function  $A(\Delta\lambda)$  of the measuring instrument since more than one Raman line will be transmitted by the device. The apparatus function, with a peak transmission at wavelength  $\lambda_p$ , may then be scanned through the spectrum. The unresolved spectral intensity  $N_{\lambda_p}$  is then given by:

$$N_{\lambda_p} = \sum_{v,J} N_{v,J} A(\Delta\lambda) \quad (13)$$

and

$$\Delta\lambda = \lambda_p - \lambda_{v,J} \quad (13)$$

where

$$\lambda_{v,J} = \frac{10^8}{\nu_0 - \Delta\nu_{v,J}}$$

$N_{v,J}$  and  $\Delta\nu_{v,J}$  are the Raman line intensities and frequency shifts, respectively, given by the detailed equations in the previous section. By varying  $\lambda_p$  through the region of the Raman lines, the unresolved spectral scan can be determined theoretically using a digital computer program to perform the straightforward but tedious computations. The computer code does nothing more than take each of the few thousand Raman line intensities and sum the product of these intensities and the apparatus function transmission values at the Raman wavelength of the particular transition. Each sum corresponds to a particular wavelength position of the peak of the apparatus function (or transmission curve if you will). The computer then increments this peak wavelength by a specified amount and repeats the entire procedure again and again until a theoretical wavelength scan is produced. Most of the Raman lines will not add to the sum of a narrow apparatus function because the transmission is zero at their particular wavelength, also when  $\lambda_p$  is outside the range of the larger Raman intensities, the sum will be negligible even at the peak transmission of the apparatus function. An example of such a calculation for molecular nitrogen at a constant number density and temperatures of 300°K and 3000°K is shown in Figure 2 for a ruby laser source where  $\lambda_0 = 6943\text{\AA}$ . A narrow 5\text{\AA} half-width triangular apparatus function was assumed. Note that individual rotational lines are not detectable as in Figure 1 since the apparatus function is wide enough to include many lines at once. However, the various Q-branches are able to be resolved with this particular apparatus function. Also, note the vast difference in the spectra at the two temperatures shown in Figure 2. At 300°K the Raman spectrum is limited to a narrow region of the total spectrum while at 3000°K a much broader wavelength interval is covered.

Since most early Raman work was done at room temperature or below, the term Raman "line" was adopted, because the Raman spectrum looked like a "line" (e.g., see Figure 2). However, approximately 20 actual rotational lines make up the Q-branch spectrum at room temperature, as shown in Figure 1; and, therefore, the term "line" is incorrect. At higher temperatures, where other Q-branches appear due to upper level vibrational transitions, the term "line" becomes

meaningless as well as incorrect. Therefore, a better term would be Raman "rotation-vibration spectrum" or just "Raman spectrum," which has been adopted in this report.

The apparatus function used in the calculations plotted in Figure 2 was somewhat narrow for the spectral analyzer used in this study. Therefore, the actual apparatus function employed in the present experiment was used to compute the unresolved Raman spectrum. The results of these calculations are shown in Figure 3 for oxygen at temperatures of  $T_v = T_r = 300, 900$  and  $1500^\circ\text{K}$ . Note that the various Q-branches are not resolved for this relatively broad apparatus function, yet the spectral intensity profile is quite sensitive to gas temperature.

For a particular  $\Delta v=1$  transition (i.e., (0,1) O-branch), Eq. (4) can be written:

$$\frac{N_{0,J}}{N_{0,0}} = \left[ \frac{\nu_0 - \Delta\nu_{0,J}}{\nu_0 - \Delta\nu_{0,0}} \right]^4 \frac{S_J g_J}{S_0 g_0} e^{-\frac{1.44 F(J)}{T_r}} \quad (14)$$

Thus, the relative line intensity distribution in a given O-branch is a function only of the rotational temperature of the gas. This can be readily observed in Figures 1 and 2, where the wavelength of peak intensity in the (0,1) Q-branch has shifted toward the blue end of the spectrum at the higher temperature condition. Also, note that the asymmetry of the spectral profile is quite different for the elevated temperature case. Thus, by curve fitting the spectral profile of the (0,1) Q-branch and/or any other branch, the rotational temperature of the gas can be obtained from the rotation-vibration spectrum. Since the branches must be resolved, the narrower apparatus function, such as that used in Figure 2, must be used in order to obtain information on the rotational state of the scattering molecules.

Summing Eq. (4) over the rotational quantum number  $J$  will result in the following expression:

$$\frac{N_v}{N_0} = \left[ \frac{\nu_0 - \Delta\nu_v}{\nu_0 - \Delta\nu_0} \right]^4 S_v e^{-\frac{1.44 G(v)}{T_v}} \quad (15)$$

where

$$\Delta\nu_v = \omega_e - 2\omega_e x_e(v+1) + \omega_e y_e(3v^2 + 6v + 13/4) \quad (16)$$

Hence, the relative integrated intensity of the branches is indicative of the vibrational state of the gas. In other words, the shape of each branch is a function of the rotational temperature, but the relative intensity of the various branches is dependent only on the vibrational temperature of the scattering molecules. It must be pointed out that, in general, accurate integrated Q-branch intensities are difficult to obtain due to the overlap of the various branches. Thus, Eq. (14) cannot be used in most cases to calculate the vibrational temperature from the unresolved spectrum. Instead, curve fitting techniques must be employed to obtain the vibrational temperature of the gas. Note that both of the above spectral analysis methods require only relative intensity measurements; no absolute intensities are needed.

For equilibrium conditions (i.e.,  $T_v = T_r = T$ ), a curve fit of the entire spectral intensity profile can be used to obtain the gas temperature. Either narrow or broad apparatus functions as in Figure 2 or Figure 3 can be used. However, if thermodynamic nonequilibrium is suspected, a separate curve fit must be made of one or more Q-branches to determine the rotational temperatures independently of the vibrational state of the gas. Then, the vibrational temperature can be determined from a curve fit of the entire rotation-vibration spectrum. The rotational temperature of the gas can be used as data for this calculation. Note, that narrow apparatus functions, as in Figure 2, must be used for the nonequilibrium case in order to determine independently the rotational and vibrational states of the test gas.

#### D. NUMBER DENSITY MEASUREMENT

A cursory examination of Eq. (4) might imply that the Raman spectrum could be used directly to evaluate the number density,  $n$ , of a gas under varying temperature conditions. This is not true since Raman line intensities are dependent upon temperature and number densities simultaneously. This coupling of number density and temperature is common to all spectroscopic techniques and arises from temperature effects on the population distribution of the various energy modes. The coupling effect can be readily demonstrated by considering the spectral plot shown in Figure 2, where constant number density was used in the calculations. Note that if the (0,1) Q-branch peak intensity were to be used to compute number density at 3000°K without making a correction for temperature effects, the calculated values, compared to room conditions, would be in error by a factor of about 5. Hence, the gas temperature must be known in order to obtain the correct number density of any of the species in the test gas. A temperature determination is, however, not dependent on knowledge of the actual density. For nonequilibrium conditions, both the vibrational and rotational temperatures must be obtained in order to correct for temperature dependence. This temperature effect on number density measurements can be minimized under certain circumstances. Note that by using a broader apparatus function, as shown in Figure 3, the resulting oxygen Raman spectrum

is nearly temperature independent at  $7778\text{\AA}$ . Hence, by choosing proper experimental conditions the need for temperature corrections can be eliminated if a certain amount of calculable error is tolerable. In this case, the intensity of the oxygen Raman spectrum at  $7778\text{\AA}$  is nearly directly proportional to the number density of oxygen for the particular apparatus function used in the calculations. Varying the shape of the apparatus function will change the optimum wavelength and also the error estimates.

To determine the number density of the constituents of high temperature air, the measurement of absolute spectral intensities is not required. Those difficult absolute measurements may be avoided through the application of a calibration point which is the known composition and number density of room air existing before and/or after the high temperature test condition. Also, the number density of any species which is present only in the test environment can be determined by ratioing its intensity value to a room temperature constituent such as nitrogen. Of course the nitrogen would also have to be present in the test environment. This is an attractive feature of the Raman scattering diagnostic method over other spectroscopic techniques which require either absolute intensity measurements or complex calibration procedures.

## E. RATE MEASUREMENTS

### 1. Vibrational Excitation Time

A finite time is required for the vibrational temperature to equilibrate with temperatures of the rotational and translational modes of motion during rapid changes in molecular energy. This nonequilibrium process can be investigated using Raman scattering. To monitor both the vibrational and rotational temperature would be straightforward but cumbersome. A narrow apparatus function must be used, as discussed above, and data must be simultaneously obtained from at least three different wavelength positions requiring a good deal of instrumentation.

This type of temperature data would be informative; however, often only the time for the vibrational mode to equilibrate (i.e., vibrational-excitation time or  $\tau_{v-T}$ ) is of primary interest. A simple method can be used to determine  $\tau_{v-T}$  utilizing Raman spectroscopy by monitoring the Raman intensity using a single spectrum analyzer. In this case, the transmission peak of the apparatus must be located at the proper wavelength position in the spectrum. Raman intensity calculations were performed for oxygen using Eq. (13) and assuming an abrupt change in the rotational temperature from ambient conditions to a value of  $1100^\circ\text{K}$ , due to the passage of a shock wave. The vibrational temperature was then theoretically increased from  $300^\circ\text{K}$  to  $1100^\circ\text{K}$  (i.e., from ambient to equilibration with the rotational temperature) as occurs behind the wave during vibration excitation. Results of these calculations are shown in Figure 4. Note that there is a significant change in the spectrum with just the rotational mode excited. Then the spectrum changes further as the population



of the upper vibrational state increases. Hence,  $\tau_{V-T}$  is the time between the first change just behind the wave and the attainment of a constant value of the Raman intensity at thermodynamic equilibrium. Note that in Figure 4 the broad 22.5Å apparatus function is sufficient for these excitation time experiments.

## 2. Chemical Reaction Time

As with the internal modes of motion, chemical reactions also take a finite time to occur. Usually the excitation time for the internal modes is much shorter than reaction times so coupling can often be neglected. Measurement of the chemical reaction rate using Raman scattering involves monitoring the formation or consumption of a particular constituent with time. However, for a pure gas the chemical reaction process alters the heat content which results in a temperature change during the reaction. As discussed in Section D, spectroscopic intensities are functions of temperature as well as density. Hence, errors could be introduced into the rate measurements if this temperature-density coupling is not taken into account.

Again, one may monitor both the temperature and density. Thus, Raman intensity data would have to be obtained simultaneously for at least two different wavelength positions. The measured temperature could then be used to obtain corrected density histories from which chemical rates could be computed. A simpler way which avoids the temperature-density coupling would involve diluting the chemical reactants in a buffer gas such as argon. The chemical reaction would result in a negligible change in the total gas energy so the temperature would remain nearly constant. Now, a single measuring instrument (i.e., one broad apparatus function) could be used to directly monitor density changes with time. Chemical reaction rates could then be calculated without any error introduced by the need to measure temperature.

## III. SHOCK TUBE IN RAMAN SCATTERING EXPERIMENTS

## A. GENERAL DISCUSSION

The shock tube is essentially a device in which a planar shock wave is generated by the sudden bursting of a diaphragm which separates a high-pressure low molecular weight gas from a test gas at low pressure. Behind this wave the experimental gas is brought almost instantly to a known and controlled high temperature, held at steady temperature and pressure for a few hundred microseconds, and then cooled suddenly. If the gas is studied at some point along the tube, then it is possible to follow the progress of chemical reactions, and vibrational excitation processes as the sample of heated gas passes the measurement point. Since the density of the gas behind the wave is usually quite high, then Raman scattering measurements are facilitated by the increase in scattering intensity. The generation of a known thermochemical state of high density gas implies that the shock tube is a valuable tool for calibrating Raman measurement apparatus and validating Raman theory at elevated temperature conditions.

## B. BRIEF DESCRIPTION OF FACILITY

To generate the temperature and density conditions necessary for this study, a 25 mm I.D. 3.5 meter long shock tube was used. The driver was operated with helium while the driven tube contained either pure nitrogen or dry air. Scored aluminum diaphragms were used and they were punctured with a hand operated plunger after preset charges of gas were placed in the tube. The plunger method provided repeatable shock velocities which were essential to obtain accurate data for comparison with shock tube theory. A small dump tank was used following the test section.

Driven pressure,  $p_1$ , was measured with a 0-400 torr Wallace-Tiernan gauge and driver pressure,  $p_4$ , was accurately and repeatably preset with a pressure transducer and digital readout system. The wave speed was determined for each shot using three pressure transducers to start and stop two Monsanto Model 401 timers which were accurate to within one microsecond. The pressure transducers were located on either side of the test section and the shock speed at the point of the Raman measurements was computed assuming linear shock speed attenuation. Error estimates showed that the computed speeds were accurate to within 2 percent.

For the Raman experiments, the tube was altered in several ways. A fourth pressure transducer was added near the diaphragm. The purpose of this transducer was to provide a trigger signal to fire the laser. Another alteration was the addition of a Raman test chamber near the end of the tube. Finally a support was attached to the end of the tube and bolted to the floor. The purpose of this stiffener was to minimize any vibration during the diaphragm rupturing. Removing this vibration eliminated any significant misalignment of the beam with the collection optics.

### C. POSTSHOCK PROPERTIES

A pressure limit on  $p_4$  of 325 psia was arrived at from safety considerations since the diaphragm plunger was hand operated. Also, to obtain reasonably high postshock densities  $p_1$  was kept above 50 mm-Hg in the present experiments.  $p_4$  and  $p_1$  were adjusted accordingly to produce shock speeds from 1100-1600 m/sec. The above restrictions were not prohibitive since they provided a postshock temperature range of approximately 750-1400°K and densities from about 0.35-0.75 the density of air at STP conditions.

Another postshock parameter which had to be considered was the vibrational excitation time of a gas constituent compared to the test time of the shock tube. The excitation time had to be shorter than the test time or equilibrium conditions could not be achieved behind the wave. The test time ranged from 100-300  $\mu$ sec, therefore, only pure oxygen or  $O_2$  in air could be investigated for lower speed shocks. At the higher shock velocities, the properties of  $N_2$  in air were investigated. Pure  $N_2$  could not be examined at any equilibrium conditions in the present tube because of the long excitation times compared with the test time restrictions.

#### IV. RAMAN INSTRUMENTATION FOR SHOCK TUBE DIAGNOSTICS

##### A. GENERAL DISCUSSION

The application of Raman spectroscopy to a shock tube environment is in general a straightforward process of obtaining the required scattering data at the proper time and within a short time interval. Also, the measurements must be made in a small region within the center of the tube. The recording time must be short in order that instantaneous and not time averaged results are obtained within the test time. When rate measurements are being made, the measurement time must also be short compared to the nonequilibrium thermochemical times associated with the shock heating process. The scattering volume must be small to avoid boundary layer regions of the flow which would complicate the calculation of postshock gas properties. A small scattering volume dimension parallel to the shock front is also required if accurate time histories are to be obtained behind the wave for thermochemical rate measurements.

The particular experimental instrumentation and associated optical apparatus necessary to fulfill the above requirements are discussed in the following sections.

##### B. OVERALL EXPERIMENTAL SETUP

A schematic of the Raman scattering apparatus for the present shock tube measurements is shown in Figure 5. Figure 6 is a photograph of the setup which emphasizes the optical components and the path of the laser beam. In Figure 7, a view of the entire experimental setup is shown which includes the spectrum analyzer and recording oscilloscope.

Since the shock tube generates high temperatures and pressures for only a few hundred microseconds and thermochemical times can be orders of magnitude less than that, pulsed lasers are best suited for Raman measurements in these transient flow facilities. Hence, a 100 megawatt pulsed ruby laser with a pulse width of 20 nanoseconds was used as the monochromatic source.

The ruby radiation at 6943 Å traveled along a path under the shock tube and in a direction parallel to the tube's axis. The beam was then rotated 90° as shown in Figure 5. Two right angle internal reflecting prisms were actually used as shown in Figure 6 to

properly turn the horizontally polarized beam and also rotate the axis of polarization so that it was in the plane of the paper as the radiation passed through the test volume. The 1 centimeter diameter beam was then focused with a cylindrical lens into the center of the 25 mm diameter tube. A 6 mm diameter spatial filter was used to obtain a 1 x 6 mm beam dimension. Intersection of this beam volume with the 0.5 x 6 mm high slit image of the spectrum analyzer defined a 0.5 x 6 x 6 mm scattering volume shown in Figure 5. Thus, the measured densities and temperatures were obtained from light scattered within the above volume. This small spatial resolution implied that good time resolution was obtained behind the shock wave and also insured that boundary layer effects were negligible. Comparison could then be made of Raman data with simple shock tube theory.

After passing through the test region, the laser radiation was collected in a light trap to avoid detection by the Raman signal processing instrumentation. A small portion of the incident radiation was first diverted with a beamsplitter to the laser power monitor which consisted of a photodiode and appropriate attenuation filters. The power monitor was required in order to record pulse-to-pulse variation in the laser output. Since the Raman signal is proportional to the incident laser intensity, this variation was ratioed out to obtain accurate Raman intensities. Note that only relative intensity measurements are required so absolute laser power measurements are not necessary. This fact will be discussed in more detail later in the report.

As shown in Figure 8, the Raman scattered light at 90° to the incident laser beam was collected with a f/2.5 lens located a focal distance of 13 mm from the scattering volume. A f/5 lens focused the collected radiation on to the slit of the spectrum analyzer.

### C. ELECTRO-OPTICAL COMPONENTS

The scattered light collected by the optics must be analyzed to determine its spectral content. In the present experiment a 3/4 meter f/6.7 Spex spectrometer was used. A 600 gr/mm grating blazed at 1μ was employed in conjunction with 1 mm slit. This combination resulted in an apparatus function which was nearly triangular with a 1/2 bandwidth of 22.5Å in the first order. The true apparatus function,  $A(\Delta\lambda)$ , was measured with a helium neon laser and the data fit with mathematical functions. These equations were coded into the Raman theoretical intensity program to predict the unresolved spectra as a function of temperature using Eq. (13).

After the scattered light was spectrally analyzed it was converted to an electrical signal by an RCA 31034A photomultiplier tube, PMT, which was selected for a high quantum efficiency of 19 percent at 8600Å. Before reaching the PMT, the light passed through a laser blocking filter. The filter used in the present study was a 3 mm thick piece of RG-N9 black glass made by Schott Optical Glass, Inc.

This filter transmitted 85 percent of the incident radiation at wavelengths greater than 7700Å (i.e., the Raman region of interest) yet blocked ruby laser radiation by a measured factor of nearly  $10^5$ . The filter was used to eliminate laser radiation which was not removed by the spectrometer. Removing this unwanted light was necessary to prevent masking of the weak Raman radiation from the test gas.

Raman signal from the PMT was recorded on channel #2 of a Tektronix 454 oscilloscope using an EG&G Model IT 100 inverting transformer to invert the negative PMT signal. A 50  $\Omega$  feed-through terminator followed the inverting transformer to provide fast response times in order to accurately record the 20 nanosecond pulses. A positive output signal from the Korad KD-1 photodiode incident laser intensity monitor was also terminated in 50  $\Omega$  and recorded on the same oscilloscope by feeding this signal into channel #1 and putting the scope in the "add" mode. Since there was approximately a 100 nanosecond transit time of the electrons through the dynode channel of the PMT, then this signal arrived after the photodiode signal. Hence, both were recorded on the same instrument. An example oscilloscope trace recorded in this fashion is shown in Figure 9. A trace of this type was obtained for each laser firing. Data points then correspond to ratioing the Raman signal,  $I_R$ , to the laser signal,  $I_L$ , taken off the same trace.

The oscilloscope sweep was triggered by utilizing the negative energy pulse which is also provided by the KD-1 photodiode circuit at the time the laser is fired.

#### D. COINCIDENCE TIMING

As mentioned above, the laser had to be fired at the proper time behind the shock front. At first the solution to this problem seemed to be a simple matter of using a single transducer to provide an output when the shock wave passed a certain point. This output could be suitably delayed to fire the laser at the proper time. However, the nature of a ruby laser firing sequence complicated matters somewhat. The ruby rod must be optically pumped with a flash lamp for approximately 1 millisecond before a giant pulse can be obtained by Q-switching the cavity. Hence, using a single trigger meant that flash lamp triggering would occur when the shock wave was only about half way down the tube. Small variations in shock speed were amplified over a long distance and a 50  $\mu$ sec jitter was encountered in the time that the laser was fired with respect to passage of the shock front. The actual position of the wave with respect to the laser firing was easily obtained to a high degree of accuracy by using the KD-1 photodiodes' negative energy pulse to start a timer which was stopped by passage of the wave by position #3 in Figure 5. However, using a single transducer, the actual measured time would be a large random scattered value around a preset time rather than the value picked by the experimenter.

To remedy this situation, one transducer located near the diaphragm (i.e., transducer DT in Figure 10) and delay was used to fire the lamp while a second transducer and delay fired the Q-switch. This second

transducer (i.e., #1 in Figure 10) was located near the scattering volume in order to obtain a jitter of only  $\pm 2$  microseconds. Other details of the timing and coincidence circuitry are shown in Figure 10. Each trigger circuit consisted of a pressure transducer and a cathode follower amplifier which was used to drive a trigger generator. The 5 volt output of the trigger generator started a preset delay. The 15 volt output of the delay provided a trigger signal compatible with the laser circuitry. Using this two trigger method, the transducer signals were suitably delayed to obtain both maximum pumping of the laser rod for efficient lasing and proper orientation of the shock front with respect to the viewing port. The Q-switch transducer was also used to start two Monsanto counter timers for shock speed measurements as mentioned above.

## V. SHOCK TUBE RAMAN DATA

## A. TIME HISTORY OF SCATTERING INTENSITY

Raman scattering intensities were monitored as a function of time as a shock wave passed the viewing port. Incident laser intensities were also recorded and  $I_R/I_L$  ratios were obtained. Results of these measurements are shown in Figure 11. Each data point corresponded to a single firing of the shock tube. The laser was also fired once with each shot but with a variable delay. Hence, the first portion of the data (i.e., open circles) at  $\lambda = 8280\text{\AA}$  in Figure 11 was scattering from nitrogen ahead of the wave at the initial driven gas conditions (i.e.,  $\rho = \rho_1$ ). The final data points correspond to scattering from shock heated nitrogen. At 62  $\mu\text{sec}$  before the wave passes position #3 (see Figure 5), the Raman scattering intensity increases abruptly. This increase represents the passage of the wave through the scattering volume. The constant intensity conditions behind the wave lasted for approximately 150  $\mu\text{sec}$ . After that time a second abrupt change in scattering intensity occurred. This second increase was most likely due to passage of the contact surface through the scattering volume. The measured test time compared quite closely to the estimated value for the experimental conditions listed in the figure.

To insure that the output in Figure 11 was not some false signal, the intensity at  $\lambda = 8310\text{\AA}$  was also measured as a function of time (i.e., square symbols in Figure 11). Note that negligible output was obtained at this wavelength which was just outside the  $\text{N}_2$  Raman region. Hence, the recorded output at  $8280\text{\AA}$  was truly due to Raman scattering from nitrogen.

Also shown in Figure 11 (i.e., diamond symbols) is the amount of scattering obtained from 600 torr of nitrogen at static conditions. This pre-shot data was used as a calibration bench mark to determine the absolute density prior to and behind the wave. The data in Figure 11 was reduced to true density ratios by dividing each data point by the average intensity value of  $I_R/I_L = .13$  ahead of wave. The data was also corrected for temperature effects as discussed in Section II-D. This temperature correction will be further clarified in the following sections. The measurements in Figure 11 were also adjusted by subtracting 62  $\mu\text{sec}$  from the recorded times. The results of this entire data reduction process are shown in Figure 12. Also shown in the figure is the theoretical plot for density ratio based on constant ratio of specific heat of  $\gamma = 1.4$ . This theory was picked



because, for pure nitrogen under the present experimental conditions, the vibrational mode would not be excited in the short test time encountered in this experiment. Note that the measured ratio was 10 percent greater than the theoretical prediction for this particular case. This disagreement was reasonable considering that the data in Figure 12 were, to the author's knowledge, the first Raman measurements behind a shock wave and much was learned about improving the experimental procedure following this initial data gathering sequence.

#### B. DETERMINATION OF VIBRATIONAL EXCITATION TIME FOR OXYGEN IN AIR

As discussed in Section II-E, the vibrational excitation time for a gas can be determined by examining the Raman scattering profile as a function of time behind the wave. This time history data was obtained for oxygen vibrational excitation in air at a shock velocity of approximately 1100 meters/sec and initial air pressure of 150 torr. The data consisted of measuring the ratio of the Raman scattering intensity at  $\lambda_1 = 7762\text{\AA}$  to the incident laser intensity at various times relative to the passage of the shock front. The results are shown in Figure 13. Also plotted for illustrative purposes in the figure are the constant density profile shapes for various shock conditions and the positions on the time history plot where these conditions prevail. Note that the first profile represents static gas at ambient conditions ahead of the wave. The Raman profile for this condition is symmetric indicating negligible upper level rotational or vibrational excitation. The second condition labeled (2) on the plots is indicative of the state of the gas immediately behind the shock wave. Here the rotational temperature has increased to at least the equilibrium value behind the wave while the vibrational temperature still remains at the low ambient value. The increase in rotational temperature results in a change in the profile to a more asymmetric shape as shown in the figure. The asymmetry is caused by the increased population of the upper rotational states of the oxygen molecule. Note that the increase in Raman intensity between (1) and (2) appears to be only a factor of 3 due to the profile changes alone. However, the data indicates a change closer to a factor of 12. The apparent difference between these two factors is explained by the fact that the density also increases behind the wave by a factor of 4.

Finally, the vibrational mode excitation increases with time behind the wave until the third condition is reached. This final state represents fully excited vibration and rotation which results in a further asymmetry in the Raman profile.

This final Raman intensity increase at  $\lambda_1$  was approximately a factor of 2 as noted from the theory and experimental data. The time between condition (2) and (3) is defined as the vibrational excitation time for oxygen in air.

Also shown in Figure 13 is the theoretical Raman intensity ratio represented by the solid line. Note that a dashed line was used between (2) and (3) to indicate that this path is not being exactly represented.

The prediction would be quite complex for several reasons. There would actually be a rotational temperature overshoot occurring just behind the wave and the exact vibrational temperature and rotational temperature would be difficult to predict. In actuality, there is no real need to know the exact path because only the end points are required for this excitation time measurement. In Figure 13 the theory has been implicitly corrected for temperature effects by accounting for profile changes with temperature. In fact, the existence of temperature effects is the reason behind the ability to measure the vibrational excitation process.

Note that the measured Raman intensities at the end points agree quite well with the predicted values. However, the measured time of 36  $\mu\text{sec}$  shown in Figure 13 is 30 percent greater than the theoretically predicted value of 27  $\mu\text{sec}$ .

The theoretical excitation time was computed utilizing experimental results determined in other shock-tube experiments using interferometric techniques. First the times had to be converted to lab fixed coordinates from wave fixed values since the Raman measurement was in the lab fixed coordinate system. To convert from one system to another the following formula is used:

$$t_{\text{Lab}} = \frac{p\tau_w}{p_2(\rho_2/\rho_1)} \quad (17)$$

where  $t_{\text{lab}}$  and  $\tau_w$  are the lab and wave fixed values,  $p_2$  is the post-shock pressure and  $(\rho_2/\rho_1)$  is the density ratio across the shock.  $\tau_w$  for oxygen excitation in air (i.e.,  $\tau_{\text{O}_2\text{-Air}}$ ) is given by:

$$\frac{1}{p\tau_{\text{O}_2\text{-Air}}} = \frac{\eta_{\text{O}_2}}{p\tau_{\text{O}_2\text{-O}_2}} + \frac{\eta_{\text{N}_2}}{p\tau_{\text{O}_2\text{-N}_2}} \quad (18)$$

In Eq. (18)  $\eta_i$  is the mole fraction of gas  $i$  in air.  $p\tau_{\text{O}_2\text{-O}_2}$  and  $p\tau_{\text{O}_2\text{-N}_2}$  are given by:

$$p\tau_{\text{O}_2\text{-O}_2} = 10^{56} (T^{-1/3} - .03)^{-8} \text{ [atm-sec]} \quad (19)$$

$$p\tau_{\text{O}_2\text{-N}_2} = 1.4 p\tau_{\text{O}_2\text{-O}_2} \text{ [atm-sec]} \quad (20)$$

Eq. (19) was obtained from shock tube data by White and Millikan.<sup>15</sup>  
 Eq. (20) was obtained from observations of Blackman<sup>16</sup> showing that  $N_2$  is 40 percent as effective as  $O_2$  in the V-T excitation of  $O_2$ .

#### C. TEMPERATURE AND DENSITY DETERMINATION USING SPECTRAL SCAN METHOD

As discussed in Sections II-D and II-E, the thermochemical state of a gas can be determined by measuring its Raman scattering spectrum. This intensity versus wavelength data can then be used to obtain the temperature and density of various species in a gas mixture. In this section, the postshock temperature and density measurements of  $O_2$  and  $N_2$  in air are discussed.

The objective was to determine equilibrium gas properties and compare the Raman measurements to equilibrium theoretical calculations. To insure that indeed local thermodynamic equilibrium (LTE) did exist during the test time, calculations were made of the vibrational excitation times and these computed values were compared to the expected test time in this particular experiment. As discussed in Section III-D, and V-A, pure  $N_2$  could not be examined under equilibrium conditions due to its slow excitation time. However, calculations showed that  $O_2$  in air was sufficiently rapid such that equilibrium conditions could be obtained over a wide range of conditions. Also,  $N_2$  in air could be examined at higher shock velocities due to the enhanced rate of the  $N_2$  V-T process in the presence of oxygen. For LTE conditions, the temperatures of the various modes are equal to the translational temperature of the gas (i.e.,  $T_r = T_v = T$ ). As discussed at the end of Section II-C, a broad apparatus function can be employed in this case. The following data was thus obtained using the measured apparatus function with a width at half height of 22.5Å.

To obtain accurate Raman temperature and density measurements using LRS in shock tube experiments, various precautions had to be observed during the data taking sequence. The first step was to make sure that the scattering volume was properly aligned with the spectrometer slit. This was accomplished by translating the f/2.5 lens in Figure 8 in a direction perpendicular to the slit height. This adjustment traversed the narrow 1 mm beam image across the 1 mm slit width. Raman intensity at a fixed wavelength and pressure was then obtained as a function of lens position prior to a data gathering sequence. The position of peak scattering intensity was found and the lens location set at that position. The intensity versus position profile was relatively flat near the peak value. However, periodic checks showed that small adjustments were required to maintain proper alignment. The stiffener which was added to the tube helped to reduce misalignment problems.

<sup>15</sup>White, D. R. and Millikan, R. C., "Vibrational Relaxation in Air," AIAA Journal 2, pp. 1844-1846, October 1964

<sup>16</sup>Blackman, V., "Vibrational Relaxation in Oxygen and Nitrogen," J. Fluid Mech. 1, pp. 61-85, May 1956

The next step was to obtain a Raman spectral scan at static conditions for the particular gas to be measured in the postshock state. This data consisted of ratioing Raman scattering intensities to incident laser intensity values (i.e.,  $I_R/I_L$ ) for various wavelengths throughout the Raman region. The results of such a scan for nitrogen in air are shown on the upper plot in Figure 14. This raw data was obtained by evacuating the tube to a pressure of 200 torr of air prior to a given testing period. Each data point (i.e., open symbol) corresponds to a single laser firing but at a different indicated spectrometer wavelength position,  $\lambda_I$ . Note that some residual intensity not associated with Raman scattering was present outside the  $N_2$  Raman region (i.e.,  $I_{BC} = .2$ ). This background light was probably due to photons produced from fluorescence of the liquid absorber used in the laser dump. The measured Raman intensities had to be corrected for this effect by subtracting  $I_{BC}$  from the data in Figure 14 to obtain  $I_C$ , the true calibration intensity. After this background light subtraction was accomplished, the data was normalized with the peak intensity value,  $I_{maxC}$ , using the following formula:

$$\bar{I} = \frac{(I_R/I_L) - I_{BC}}{I_{maxC}} = \frac{I_C}{I_{maxC}} \quad (21)$$

Note, the value of  $I_{maxC}$  in Eq. (21) was obtained by drawing a smooth line through the data points (i.e., in Figure 14,  $I_{maxC} = 2.30 - .20 = 2.10$ ). The purpose of obtaining the normalized static scan may not be obvious at this time. However, to this point in the data gathering sequence, the discussion has centered around indicated spectrometer wavelength positions,  $\lambda_I$ . These values are not a priori equal to the actual wavelengths appearing at the exit slit of the spectrometer. In fact, it would be fortuitous if they were. A wavelength calibration procedure using standard lamp sources was helpful in determining an approximate value of the wavelength correction,  $\Delta$ , given by:

$$\Delta = \lambda_I - \lambda_A \quad (22)$$

Here  $\lambda_A$  is the actual wavelength position while  $\lambda_I$  is the position that is indicated on the wavelength dial of the instrument. But,  $\Delta$  is a small but non-negligible function of  $\lambda_A$  itself so the calibration source wavelength would have to be located almost identical to the Raman wavelength. This was not practical for the present experiment, so, instead, the Raman data from Eq. (21) at known static conditions was used to accurately determine the  $\Delta$  to be used to correct the spectrometer wavelength settings.  $\Delta = 20\text{\AA}$  was determined from the normalized form of the data in Figure 14. For purposes of comparing pre- and postshock data, the static Raman output in Figure 14 was further reduced to values indicative of initial preshock conditions

(i.e.,  $p = p_1$ ). This was accomplished by multiplying the intensities that were corrected for background light by a ratio of the initial pressure,  $p_1$ , to the pressure at which the static scan data was obtained (i.e.,  $p = p_{\text{calib}}$ ). Hence:

$$I_{1M} = I_C \times \frac{p_1}{p_{\text{calib}}} \quad (23)$$

Where  $I_{1M}$  are the measured static Raman scan values referenced to the initial preshock conditions. The results of this data reduction are shown as open circles in Figure 15.

To compare this static scan data to a theory, Raman spectral intensities were computed using Eq. (13) at  $T = 296^\circ\text{K}$ . The 22.5Å experimentally determined apparatus function was used in these calculations. The computer output intensities,  $\bar{I}_R$ , are normalized to give a peak intensity of 1. These values were then adjusted to the preshock experimental conditions using:

$$\bar{I}_{1T} = \bar{I}_R (T = 296^\circ\text{K}) \times \frac{p_1}{p_{\text{calib}}} \times I_{\text{max}C} \quad (24)$$

The theoretical Raman profile at the initial static preshock conditions,  $I_{1T}$ , is the solid curve plotted in Figure 15.  $I_{1T}$  was plotted in this figure to illustrate that the computer calculation compared well with room temperature data and also to show that the value of  $\Delta$  is reasonably accurate.

Having performed the necessary alignment and wavelength calibration, the raw shock tube data was then obtained. The procedure was the same as for the static scan data except that the passage of the shock front triggered the laser to fire at the proper time behind the wave. In the present experiment this time was taken as the value after the measured excitation time and before the useful test time. The results are shown on the lower plot in Figure 14 for shock heated nitrogen in air. The postshock scan data must again be corrected for background light by subtracting  $I_{BS} = .27$  from the measured Raman to incident laser intensity ratios in Figure 14 to obtain  $I_S$ , the tube scan intensity.  $I_S$  versus wavelength data is then plotted in Figure 15 using open square symbols. The postshock measurements can now be compared to static data at preshock conditions.

To relate this scan data to actual density and thus to obtain density ratios, an accurate standard calibration is required (i.e., more accurate than the static scan data previously obtained). Such a calibration simply involves measuring the Raman scattering intensity at a known pressure (i.e.,  $p = p_{\text{STD}}$ ) at ambient temperature conditions.

Furthermore this standard intensity,  $I_{STD}$ , should be measured at a fixed wavelength location (i.e.,  $\lambda_A = \lambda_{STD}$ ).

At this point it would be appropriate to say something about the magnitude and meaning of the  $I_R/I_L$  values found in Figure 14. Note that in Figure 9,  $I_R/I_L = 2.27$  was obtained. The point to be made here is that this number is completely arbitrary and the absolute ratio need not be determined as long as nothing unknowingly happens to alter the sensitivity of the intensity monitoring equipment between the shock tube tests and the standard calibration sequence. To make sure the sensitivity is not changing with time, the standard calibration test should be made periodically between sets of shock tube firings. If this relative Raman calibration output (i.e.,  $I_{STD}$ ) at a known ambient condition is constant with time during the shock heated tests, then the sensitivities have remained constant and the calibration intensities can then be used to obtain exact results from the arbitrary intensity values shown in Figures 14 and 15. The average calibration intensity value obtained prior to and during the shock tube data sequence is also plotted as 2.18 in Figure 14 (i.e., solid circular symbol) along with the standard deviation bars representing scatter about this value. Again this calibration must be corrected for background light intensity by subtracting  $I_{PC}$  from the average value to obtain the actual standard calibration intensity (i.e.,  $I_{STD} = 2.18 - .20 = 1.98$ ). Also note from Figure 14 that the average intensity value fits quite well with the raw scan data obtained previously.  $I_{STD}$  was then altered to reflect preshock conditions so it could be used to determine the density ratio across the shock. Hence, the standard intensity at state ① conditions was given by:

$$I_{1STD} = I_{STD} \times \frac{P_1}{P_{STD}} \quad (25)$$

(i.e.,  $I_{1STD} = 1.98 \times 1/4 = .495$  from Figure 14)

where:  $P_{STD} = P_{calib} = 200$  torr

Note that Eq. (25) represents the same correction procedure that was performed on the scan data with Eq. (23).  $I_{1STD} = .495$  is then plotted in Figure 15 for comparison to the static scan data and also to reduced values of postshock data. The standard wavelength,  $\lambda_{STD}$ , was 8282Å.

The postshock Raman output in Figure 14 was then curve fit in order to obtain the temperature and density of nitrogen in the shock heated air. The trial curve fits produced residual error sums whose magnitude is based on how well the theory agreed with measured data. Two

variables were adjusted to obtain a minimum residual value. These variables were gas temperature and maximum intensity,  $I_{MAXS}$ , of the theoretical profile.  $I_{MAXS}$  was used to normalize the scan data to 1 in order to compare the measured values to the Raman theory which was also normalized to 1. Hence, Raman theory refers to results of Eq. (13) normalized to the peak intensity value. The curve fit results give postshock temperature, hence the temperature ratio, directly. However, to obtain the shock density ratio, further data reduction is required using the  $I_{MAXS}$  value obtained from the curve fit process. This further data reduction is straightforward and consists of finding the best fit value for the postshock intensity at  $\lambda_{STD} = 8282\text{\AA}$ . To accomplish this the normalized theoretical Raman spectral profile which best fit the Raman data was multiplied by  $I_{MAXS}$  to obtain  $I_T$ , where:

$$I_T = \bar{I}_R (T = T_{BEST\ FIT}) \times I_{MAXC} \quad (26)$$

Results of the calculations using Eq. (26) are then plotted for comparison with the postshock data in Figure 15. Also, the value of  $I_T$  at  $8282\text{\AA}$  was found to be 1.89 and is labeled in the figure.

At long last, the density ratio can be computed, without correcting for temperature effects, by ratioing the two intensity values at  $\lambda_{STD}$  in Figure 15. Hence:

$$\left(\frac{\rho_2}{\rho_1}\right)_{\text{uncorrected}} = \frac{I_T(\lambda=\lambda_{STD})}{I_{1STD}} \quad (27)$$

(i.e.,  $\rho_2/\rho_1 = 1.89/.495 = 3.82$  from data in the figure)

Finally, the true density ratio can be determined from the reduced data by correcting the above for temperature effects. The reason that the temperature was not accounted for in the previous procedures is that the theoretical computations used above were always normalized to a peak intensity of 1 for convenience sake in performing the curve fit process. However, as Figure 3 depicts, the peak intensity (and hence all other intensity values) varies as a function of temperature at constant density. Hence, this temperature effect must be accounted for in the data reduction process. This was accomplished by multiplying the uncorrected value by a ratio of the theoretical Raman output at  $\lambda = \lambda_{STD} = 8282\text{\AA}$  for room temperature (i.e., calibration temperature) conditions to the theoretical output at the postshock temperature. Hence:

$$\frac{\rho_2}{\rho_1} = \left(\frac{\rho_2}{\rho_1}\right)_{\text{uncorrected}} \times \frac{I_R(T=T_{ROOM})}{I_R(T=T_{BEST\ FIT})} \quad (28)$$



The theoretical Raman intensity as a function of temperature,  $I_R(\lambda) = N\lambda_p$  in Eq. (13), is plotted in Figure 16 for  $\lambda = \lambda_p = 8282\text{\AA}$ . From the figure, at  $T = T_{\text{BEST FIT}}$ ,  $I_R = .743$  and at  $T = 296^\circ\text{K}$ ,  $I_R = 1$ . Using Eq. (28), the temperature correction follows directly (i.e.,  $\rho_1/\rho_2 = 3.82 \times 1/.743 = 5.14$ ). This value is 26 percent higher than the uncorrected density ratio which indicates the importance of the temperature correction process.

The above data reduction procedure seems laborious and complex. Its description could have been simplified somewhat but a lot of the details and some of the physical meaning of the data would have been lost. Basically, Figure 14 represents the raw data containing arbitrary but consistent output sensitivities between calibration and test conditions. The raw data was corrected for background light and then referred to the initial conditions prior to the shock wave (i.e.,  $p = p_1$ ). In Figure 15, the pre- and postshock arbitrary intensity data were compared on a single plot. Temperature was obtained by a simple curve fit of the arbitrary intensity values. The density determination was more involved. Accurate pre- and postshock intensities at  $\lambda = \lambda_{\text{STD}}$  were ratioed to obtain the uncorrected density ratio. This ratioing of arbitrary intensities results in a real density ratio and hence actual postshock density simply because the preshock arbitrary intensity corresponded to a known density value. The necessary factors for correcting the density ratio for temperature effects were obtained from Figure 16.

#### D. TEMPERATURE AND DENSITY MEASUREMENT USING 2-COLOR TECHNIQUE

In many ways, the 2-color (i.e., 2-wavelength) technique is similar to the spectral scan method in that the 2-color data essentially consists of only a three point wavelength scan. Two intensity values are obtained within the Raman spectral region and a third point at a wavelength outside the Raman band in order to determine background light intensity. The idea was to obtain more accurate data at the fewer number of points by measuring Raman and background intensities at each wavelength for three to five shock tube-laser firings. The rationale behind a 2-color technique is that the spectrometer could be replaced with a series of filters and photo-detectors. This would improve the signal gathering capabilities of the optics since filter systems can have a very low f/no. And, using three filters, all the necessary experimental data to determine density and temperature could be obtained with a single laser firing.

The same precautions regarding alignment of the scattering volume with the spectrometer slit were required for the 2-color data gathering sequence. Also, a Raman spectral scan at static conditions was again necessary to convert spectrometer wavelengths to true wavelength values. The results of this scan are shown in the upper portion of Figure 17. Note that the peak intensity value of 2.51 is somewhat different than in Figure 14 indicating a small change in the sensitivity of the system in the time between the two types of data gathering sequences. The results of Figure 17 were corrected for



background light intensity and normalized using Eq. (21), where  $I_{MAXC} = 2.51 - .20 = 2.31$ . The value of  $\Delta$  determined from this static scan procedure was  $20\text{\AA}$ , which was identical to the previous wavelength correction factor.

As before,  $I_{1M}$  was obtained using Eq. (23) and the results of this data reduction procedure are shown as open circles in Figure 18. Also plotted as the solid line through this data is the computed intensity profile at the calibration temperature and pressure conditions using Eq. (24).

The standard calibration results taken prior to and during the 2-color data sequence are also shown in Figure 17 with a solid circular symbol. Using Eq. (25), the standard intensity was referred to postshock conditions (i.e.,  $I_{1STD} = (2.28 - .20) \times 1/4 = .520$ ).  $I_{1STD}$  is also plotted in Figure 18 again using a solid circular symbol.

The shock tube 2-color data was then obtained and the three intensity values are shown plotted as  $I_1 = 1.98$ ,  $I_2 = 1.33$  and  $I_B = .43$  with square symbols in the lower portion of Figure 17. The standard deviation of the data at these wavelength values is also shown in the figure using error bars. The smooth curve drawn through the postshock data points in Figure 17 is the expected Raman output and is presented for illustrative purposes only since no curve fitting is required for the 2-color technique. As the profile depicts,  $\lambda_1$  was picked near the peak of the Raman profile while  $\lambda_2$  was located in the blue shifted region of the Raman spectrum. The choice of these wavelength positions is not completely arbitrary. They were picked so that the intensity at  $\lambda_1$  would be somewhat insensitive to temperature effects (i.e.,  $I_R(\lambda_1) \approx \text{const.} \times \text{density}$ ). Further,  $\lambda_2$  was chosen so that the ratio  $I_R(\lambda_2)/I_R(\lambda_1)$  would be most sensitive to temperature changes.

From this point, the data reduction method departs rather markedly from that of the spectral scan technique. The measured postshock Raman intensities are again corrected for background light by subtracting  $I_B$  from  $I_1$  and  $I_2$  to obtain:

$$I_R(\lambda_1) = I_1 - I_B = 1.55 \quad (29)$$

$$I_R(\lambda_2) = I_2 - I_B = .90$$

The postshock 2-color intensity values in Eq. (29) are plotted in Figure 18 as solid square symbols. These two values (i.e., 2-color intensities) in conjunction with  $I_{1STD} = .523$  are all the reduced data needed to compute temperature and density using the 2-color method. Note, that for convenience,  $\lambda_1 = \lambda_{STD}$ . The solid curve drawn through the postshock data in Figure 18 represents the theoretical Raman

profile for the measured postshock temperature conditions and, again, it is only shown for illustrative purposes.

To obtain temperature from the above 2-color data, the values in Eq. (29) are ratioed to give:

$$\frac{I_R(\lambda_2)}{I_R(\lambda_1)} = \frac{.9}{1.55} = .58 \quad (30)$$

In Figure 19, the theoretical value of this ratio is computed for the particular wavelengths used in the present study (i.e.,  $\lambda_1 = 8285\text{\AA}$ ,  $\lambda_2 = 8260\text{\AA}$ ). Finding the value of the measured ratio of .58 from Eq. (30) on the vertical axis of Figure 19 and moving horizontally until the I8260/I8285 curve is intersected, one determines directly that the measured temperature is 1440°K.

Further, the uncorrected density ratio is given by an equation similar to Eq. (27) as:

$$\left(\frac{\rho_2}{\rho_1}\right)_{\text{uncorrected}} = \frac{I(\lambda_1)}{I_{1\text{STD}}} \quad (31)$$

(i.e.,  $\rho_2/\rho_1 = 1.55/.52 = 3.98$  from data in Figure 18)

Again, the true density can be determined by correcting for temperature effects. This was accomplished by multiplying the uncorrected value by a ratio of the theoretical Raman output at  $\lambda_1 = \lambda_{\text{STD}} = 8285\text{\AA}$  for standard ambient temperature conditions to the theoretical output at the measured postshock temperature. Hence:

$$\frac{\rho_2}{\rho_1} = \left(\frac{\rho_2}{\rho_1}\right)_{\text{uncorrected}} \times \frac{I_R(T=T_{\text{ROOM}})}{I_R(T=T_{\text{MEASURED}})} \quad (32)$$

This process is analogous to that of Eq. (28). The theoretical Raman intensity as a function of temperature at  $\lambda_2 = 8285\text{\AA}$  is also plotted in Figure 19. From the figure, at  $T=T_{\text{MEASURED}}$ ,  $I_R = .615$  and at  $T = 295^\circ\text{K}$ ,  $I_R = .913$ . Using Eq. (32), the temperature correction follows directly (i.e.,  $\rho_2/\rho_1 = 3.98 \times .913/.615 = 4.42$ )

## VI. DISCUSSION OF RESULTS

Excitation of pure  $O_2$  and  $N_2$ , as well as  $N_2$  and  $O_2$  in air have been discussed in this report. To clarify statements made concerning these rates, excitation times for various possible V-T processes were obtained from References 15 and 16 and plotted in Figure 20 as a function of postshock temperature. These theoretical rates are really smooth curves through data measured in shock tubes with an interferometric technique.

Pure  $N_2$  has the slowest rate, as discussed above, and it is obvious from this figure why it could not be examined in the present experiment. Also, the plot shows that the enhanced rate of  $N_2$  excitation in air is still quite slow except at higher temperatures. However, the  $O_2$  in air and pure  $O_2$  results demonstrate why  $O_2$  excitation could be examined over a larger range of conditions.

From the time history data in Figure 13, the vibrational excitation time of oxygen in air was determined at  $T_2 = 827^\circ K$ . The result was referred back to wave fixed coordinates to obtain  $(\tau_{O_2-Air})_{meas.} =$

339  $\mu sec-atm \pm 11$  percent. A theoretic value at this temperature was computed to be 250  $\mu sec$  using Eqs. (18)-(20). The Raman result is plotted in Figure 20 for comparison to the theoretical value for  $O_2$  excitation in air. The 22 percent variation in measured excitation time is an estimate based on the unknown conditions between data points in Figure 13 and the shot to shot variation in the shock speed. These shock speed variations resulted in postshock temperature and excitation time changes behind the wave.

The measured excitation time was 30 percent slower than expected. This is not an unusual variation for shock tube rate data, and more measurements at various conditions should be obtained before any firm comments on accuracy can be made. However, in defense of the Raman data, the interferometric measurements for  $O_2$  in air are restricted to temperatures above  $1500^\circ K$ . The present result is at a temperature about half that value so the disagreement may be real. Finally, no special care was taken to assure that the air used in this experiment was really dry. Bottled dry air was used, but the moisture content was never checked. Blackman's data showed that the presence of water vapor did result in a slight increase in the excitation time as was measured in this experiment.

To improve the present excitation time measurement, the ruby laser might possibly be used in its "normal" mode instead of the "Q-switched" condition. The former provides a longer pulse (i.e., 200-300  $\mu$ sec half width). Of course, the peak power falls off accordingly and one would pay the penalty of a loss in Raman scattering intensity within a given time period. However, total laser outputs of 10-20 joules are possible instead of 1-2 joules using the "Q-switch" mode. This increased energy implies that the Raman scattering output voltage on the oscilloscope need not decrease in proportion to the width of the laser pulse especially if a suitable RC time constant was used. Some further experiments would prove useful in determining if the "normal" mode could be applied successfully. The biggest advantage to "normal" mode operation would be that all the necessary data to make an excitation time measurement would be available from a single laser-shock tube firing. This can be contrasted to the 17 laser and shock tube shots necessary to obtain a single excitation time using the data in Figure 13.

The temperature and density ratio measurements obtained from the Raman technique for  $O_2$  and  $N_2$  in air at different shock speeds are shown in Figures 21 and 22, respectively. The range of postshock conditions represented by the data in the figures resulted in a temperature variation from 750 to 1400°K (i.e.,  $T_1 = 300^\circ K$ ). Absolute density variation cannot be extracted from Figure 22 directly, since variations in  $p_1$  and shock speed occurred. The actual postshock densities ranged from .35- 75 times the density of air at ambient temperature and pressure conditions.

Note that both curve fit and 2-color results, as described in the previous section, are presented in Figure 21 and 22. The vertical bars in the figures represent the maximum and minimum ratio for each data point and they indicate the extremes that could be computed due to the Raman intensity data scattered one standard deviation about the mean intensity values. The results show that the two methods of computing temperature ratios are in good agreement with each other and with the shock tube theory also plotted in the figures. The standard error is within 12 percent for temperature and 20 percent for density measurements. This disagreement between Raman data and shock tube theory is random and is mostly attributable to the uncertainty in the measured intensities. The uncertainty is mainly due to the statistical nature of the photo-electric effect in the photomultiplier tube rather than shot to shot variation in shock speed. A greater quantity of intensity data would have improved the statistical mean and thereby increased the accuracy and reduced the standard deviation scatter. Another factor that increased the scatter at the lower temperatures was the decreased sensitivity of the spectra to a temperature change. This is shown clearly when one examines the slope of the  $I_R(8260)/I_R(8285)$  ratio in Figure 19 at low temperatures.

Besides these factors, several other experimental effects, which were briefly mentioned in the text, should be reemphasized here in the discussion of the disagreement between experiment and theory.

An accurate knowledge of the actual spectrometer wavelength position during a specific Raman intensity measurement was important. The temperature and density data reduction procedure depended upon accurate  $\lambda_A$  values. Since the spectrometer wavelength was changed frequently between  $\lambda_{STD}$  and  $\lambda_1, \lambda_2, \lambda_S$ , etc. during a given data gathering sequence, errors could have resulted from improper knowledge of the true wavelength. Of course, the instrument's indicated wavelength was always set by turning the adjustment knob in the same direction to avoid gear backlash problems as one approached  $\lambda_1$ . However, hindsight indicates that a better procedure might have been to move the wavelength setting to a value about 100Å away from the desired wavelength each time it was changed. This procedure may have produced more consistently accurate  $\Delta$  values and  $\lambda_A$  settings. The wavelength inaccuracy problem may be an explanation for the larger density measurement error compared to temperature error. In the density ratio data reduction process, not only was the basic intensity data itself affected by wavelength errors, but the temperature correction factor was also highly wavelength dependent (see Figures 17 and 19). Further, since an accurate density is governed by a knowledge of the correct temperature, temperature errors compounded the density measurement problem.

Another problem was the change in sensitivity of the electro-optical system with time due to instrumentation drift and/or optical misalignment. The magnitude of this effect could be observed, somewhat, by looking at the change in the standard intensity value with time. However, it was difficult to adjust the data properly to account for sensitivity changes when purely statistical changes may have masked them. By obtaining all the data as quickly as possible, and allowing the instrumentation a sufficient warm-up time, the sensitivity drift effect was minimized but not eliminated.

The above factors probably were the largest error sources. They were much larger than the variation in shock speed from shot to shot which was no greater than 2 percent. Hence, theoretical postshock temperature and density ratios based on such shock speed variation were also less than 2 percent.

## VII. CONCLUSIONS

A comparison between the present Raman excitation measurement and data obtained from interferometric techniques demonstrates that LRS can be a valuable tool for determining shock tube excitation rates. The inherent ability of the technique to measure properties of individual gases in a general gas mixture gives it an added advantage over other shock tube diagnostic techniques, especially in the study of homonuclear molecules. Also, various molecular modes of vibrational motion in a poly-atomic molecule are Raman active and not infrared active. Perhaps some cases even exist where certain molecular modes could only be examined using the Raman scattering method. In addition to V-T excitation times, chemical reaction times, shock structure, and other shock related phenomena could be examined.

The good agreement between Raman temperature and density measurements and known values obtained from shock tube theory for a range of postshock conditions indicates several things. The LRS technique can independently determine the density and temperature of both  $O_2$  and  $N_2$  in air at elevated temperature conditions to within an acceptable accuracy. Further, this high temperature calibration experiment in conjunction with low temperature studies in air and other gases implies that utilization of the Raman scattering method should greatly enhance the amount of information which can be obtained in general 3-dimensional flow fields. The method could be applied to analysis of the flow around re-entry vehicles, in rocket exhaust, and engine plumes. In addition, LRS could be used as a diagnostic technique in laser cavities, combustors, MHD channels and wherever else one desires detailed spatial information about the thermochemical state of a flowing gas.

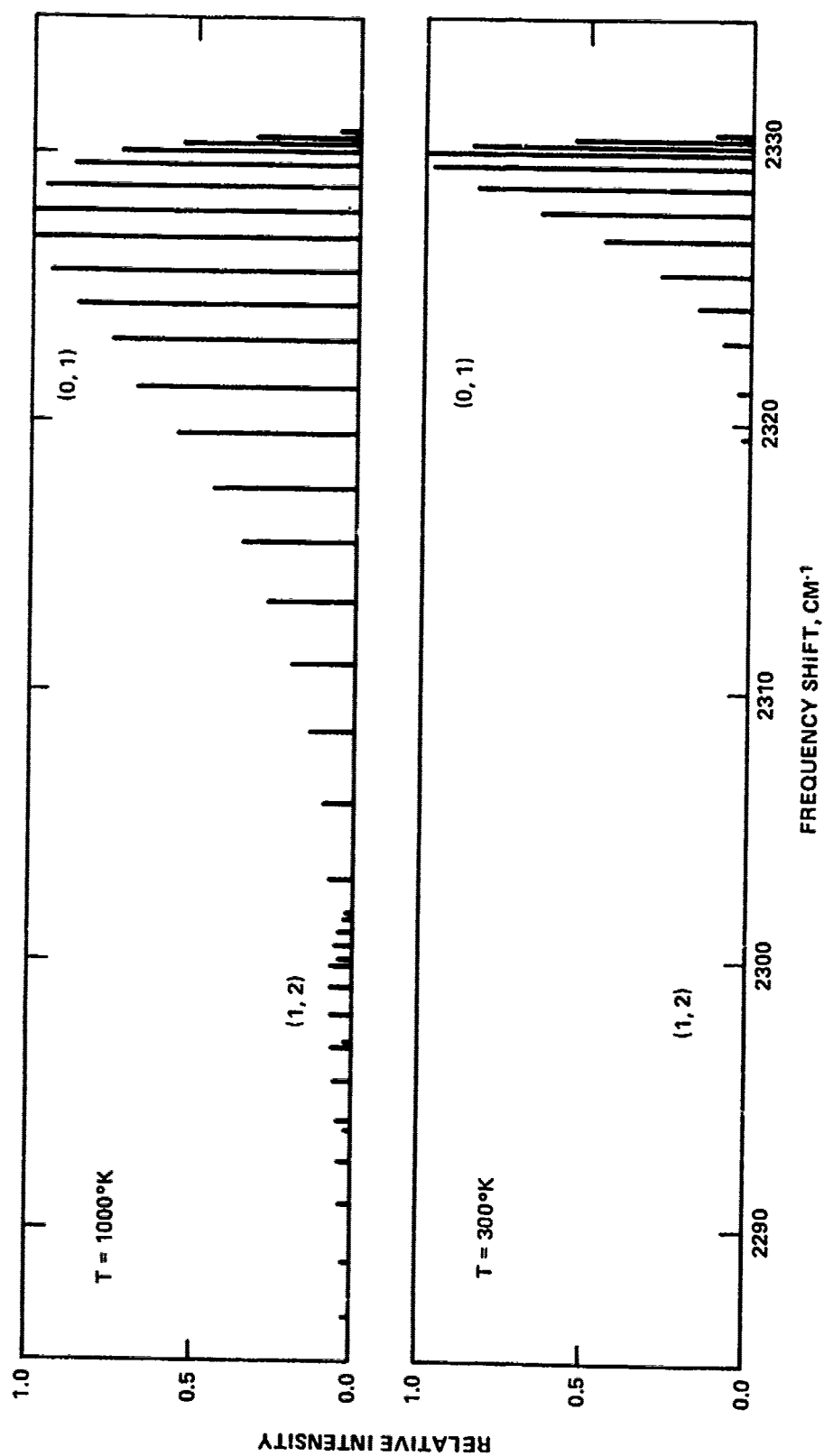


FIG. 1 CALCULATED ROTATION-VIBRATION EVEN J, Q-BRANCH LINE INTENSITIES AND FREQUENCY SHIFTS FOR MOLECULAR NITROGEN AT  $300^{\circ}\text{K}$  AND  $1000^{\circ}\text{K}$ .

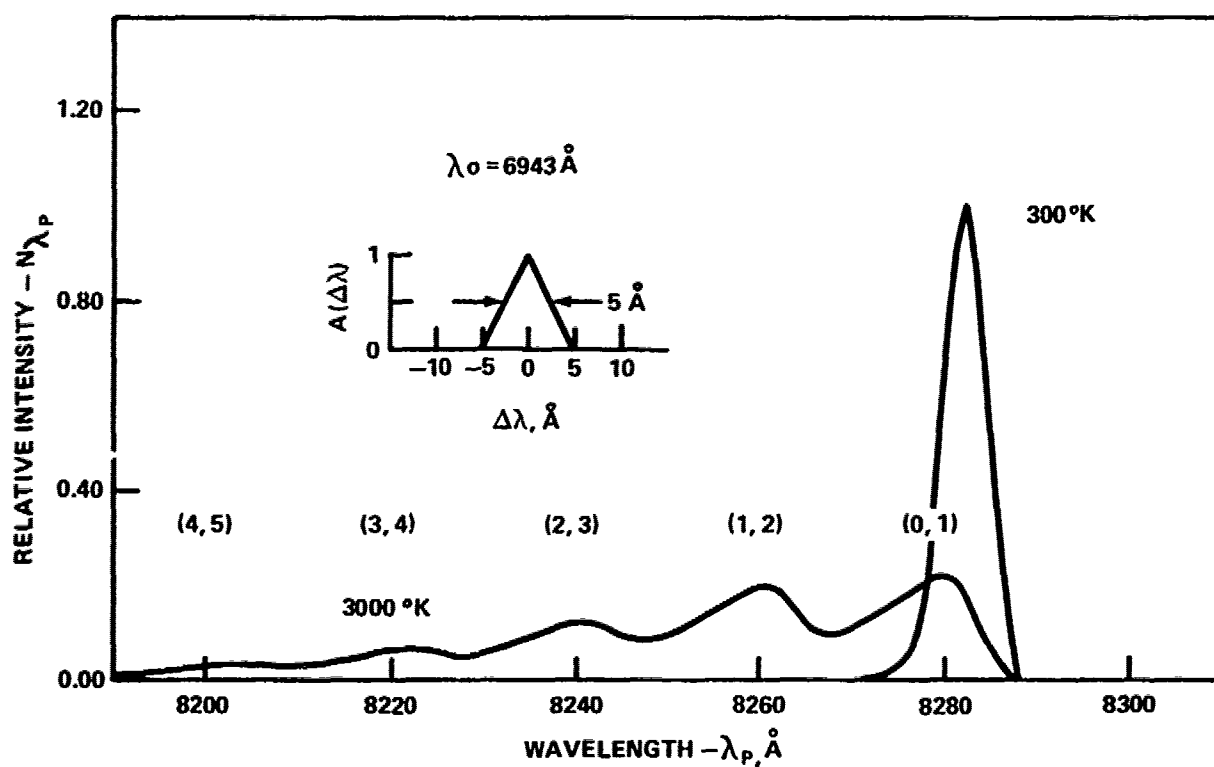


FIG. 2 UNRESOLVED STOKES ROTATION-VIBRATION SPECTRA OF NITROGEN AT  $T_V = T_R = 300^\circ\text{K}$  AND  $3000^\circ\text{K}$  COMPUTED FOR A NARROW TRIANGULAR APPARATUS FUNCTION.



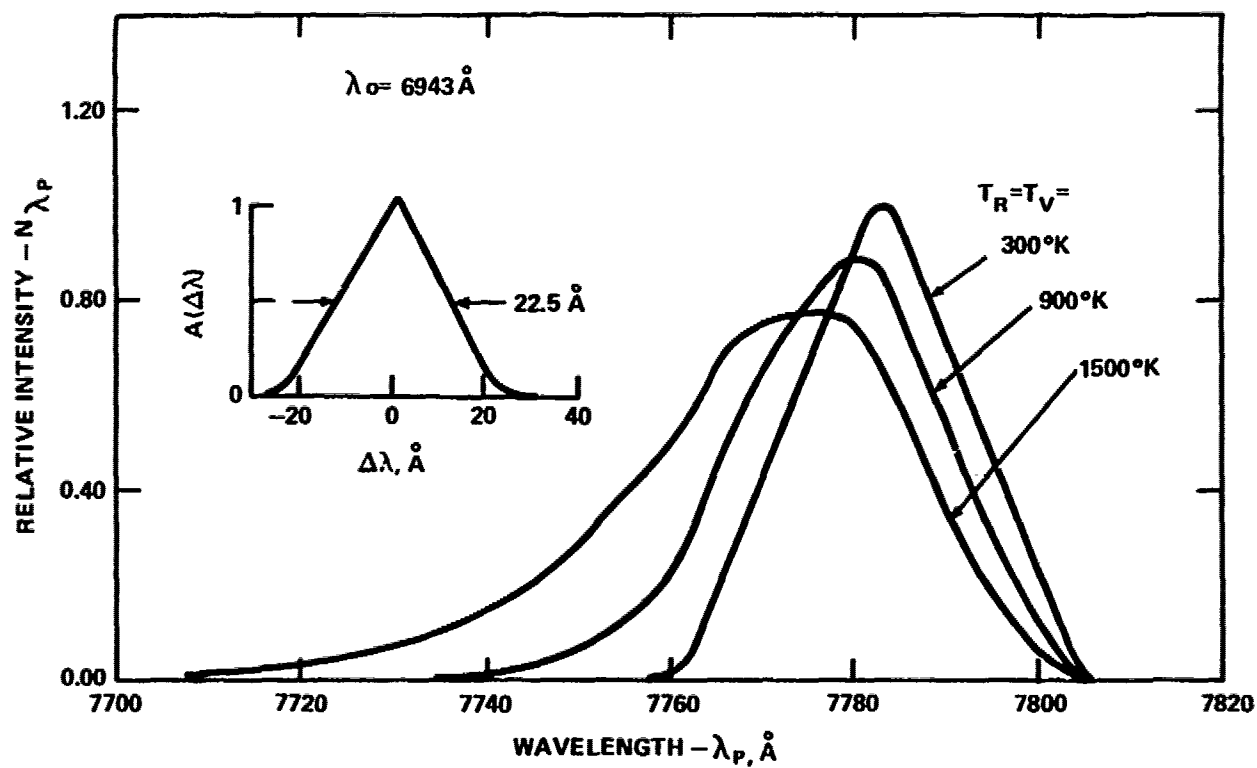


FIG. 3 UNRESOLVED STOKES ROTATION-VIBRATION SPECTRA OF OXYGEN AT  $T_V = T_R = 300^\circ\text{K}$ ,  $900^\circ\text{K}$ , AND  $1500^\circ\text{K}$  COMPUTED FOR THE BROAD EXPERIMENTAL APPARATUS FUNCTION.

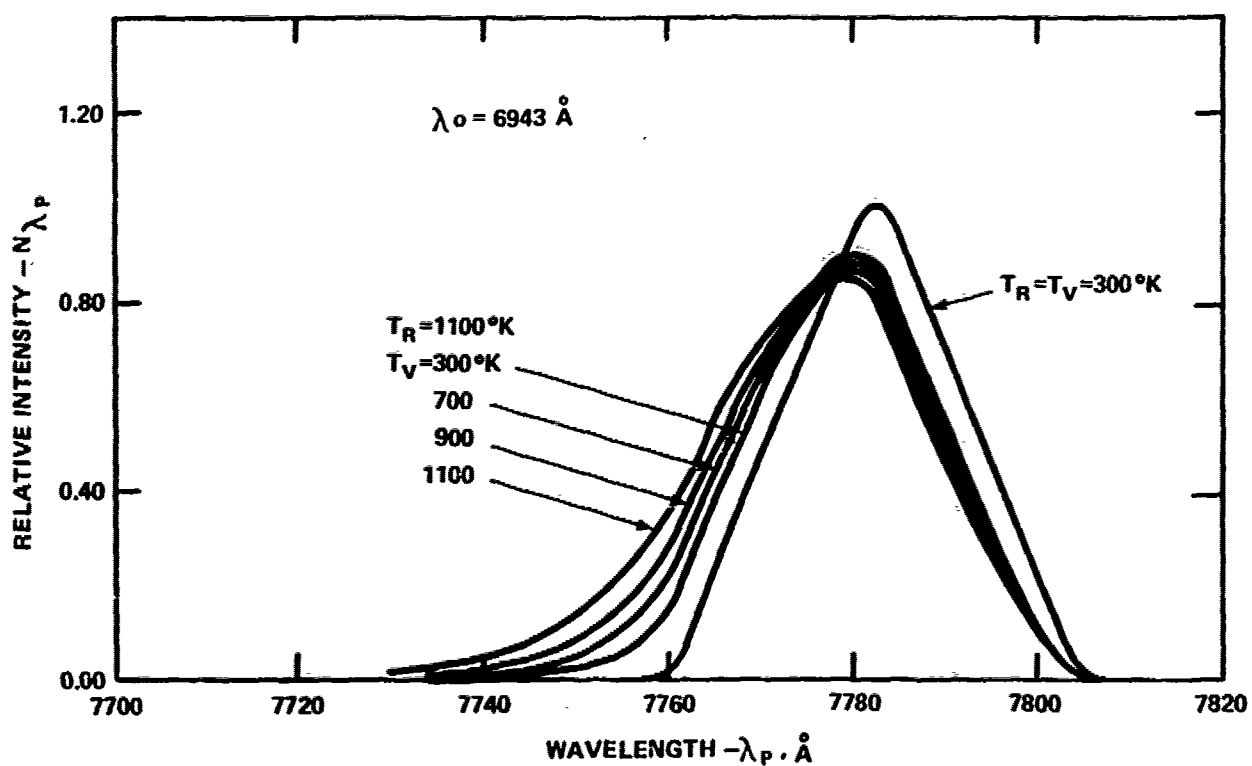


FIG. 4 UNRESOLVED STOKES ROTATION VIBRATION SPECTRA OF OXYGEN FOR NONEQUILIBRIUM CONDITIONS BEHIND AN INCIDENT SHOCK WAVE FOR  $T_V \leq T_R = 1100^\circ\text{K}$ .

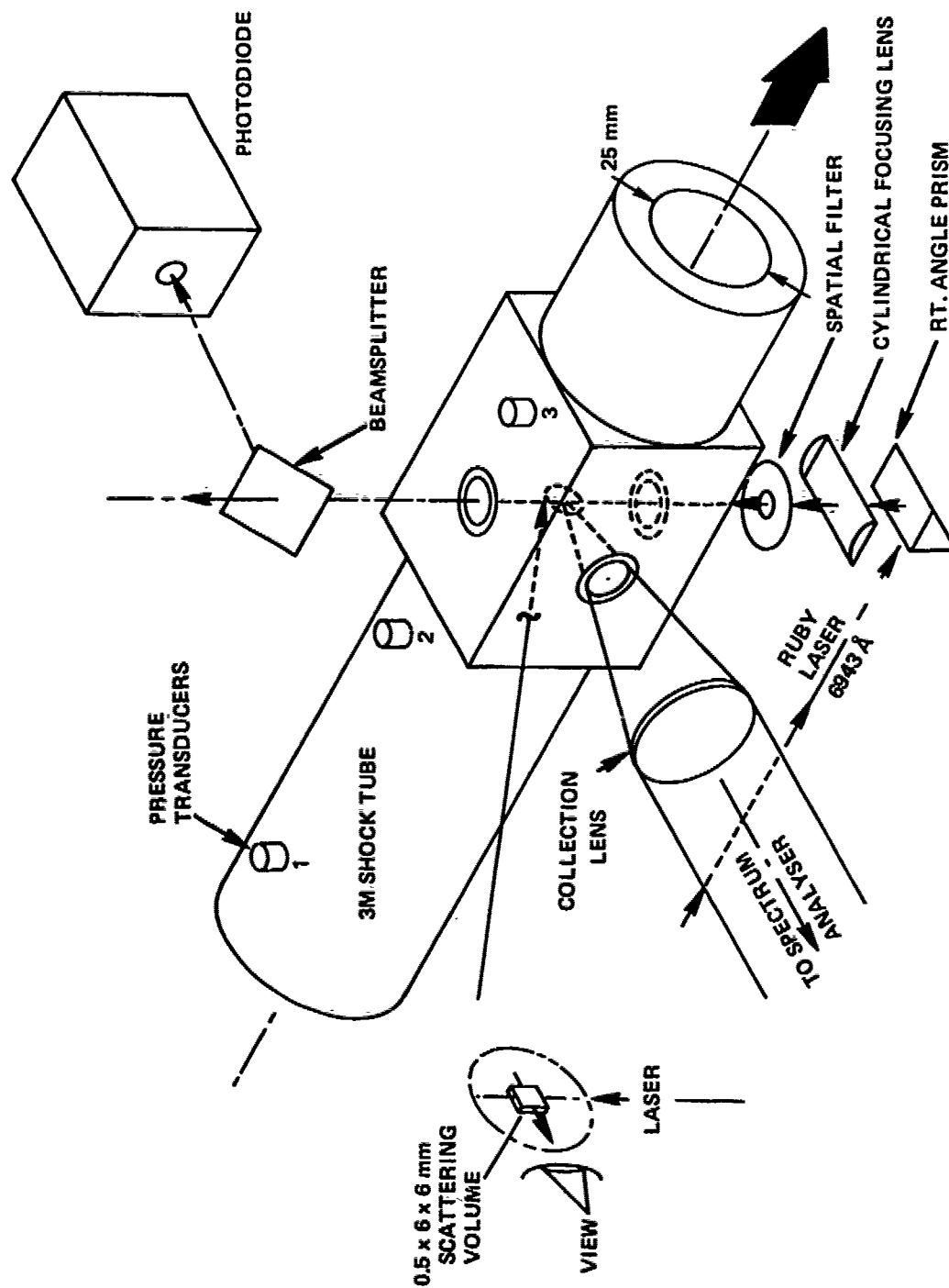


FIG. 5 SCHEMATIC OF RAMAN SCATTERING APPARATUS FOR SHOCK-TUBE EXPERIMENT

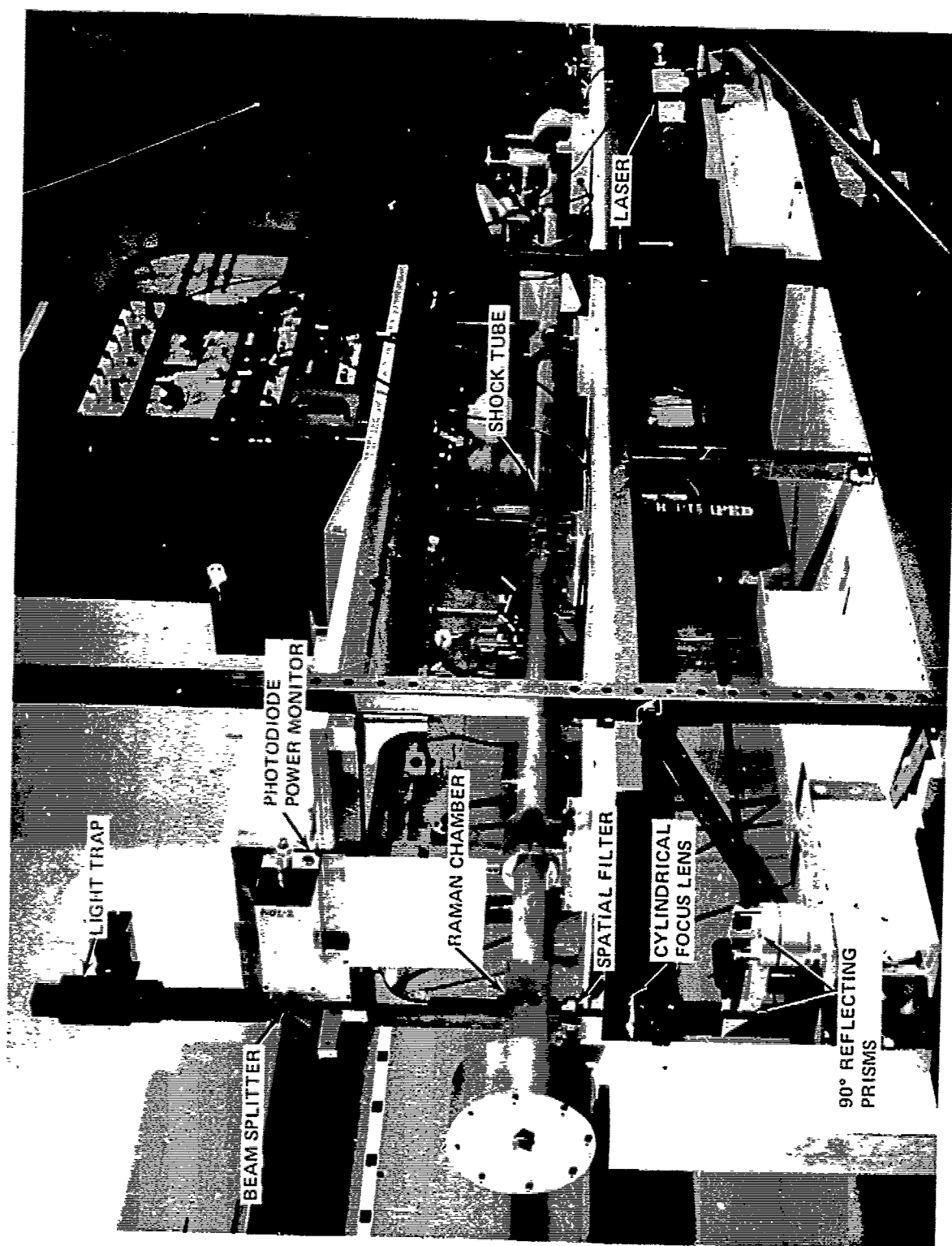


FIG. 6 RAMAN SETUP FOR SHOCK-TUBE EXPERIMENT EMPHASIZING LASER BEAM PATH

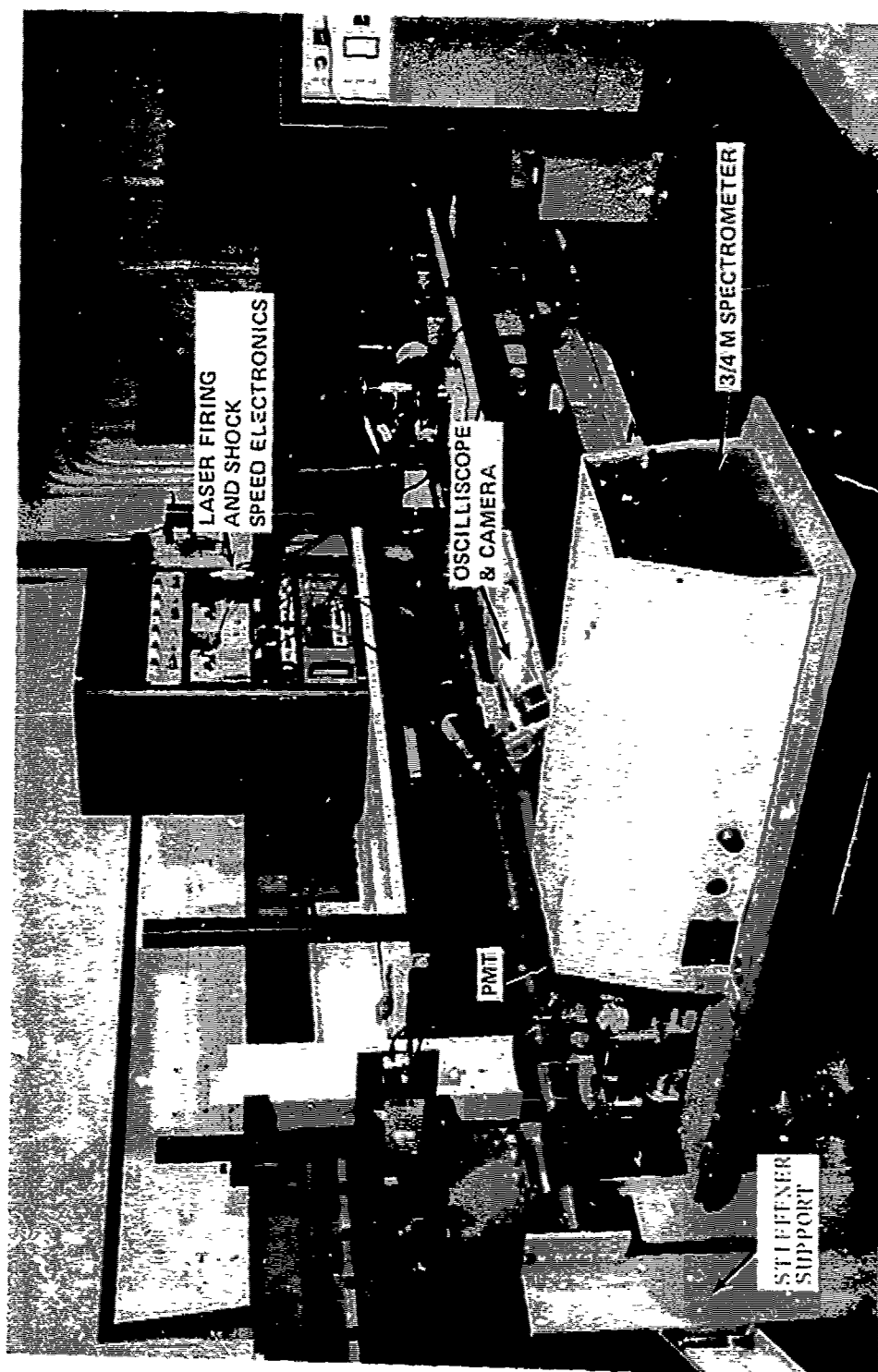


FIG.7 ENTIRE RAMAN SETUP FOR SHOCK-TUBE EXPERIMENT

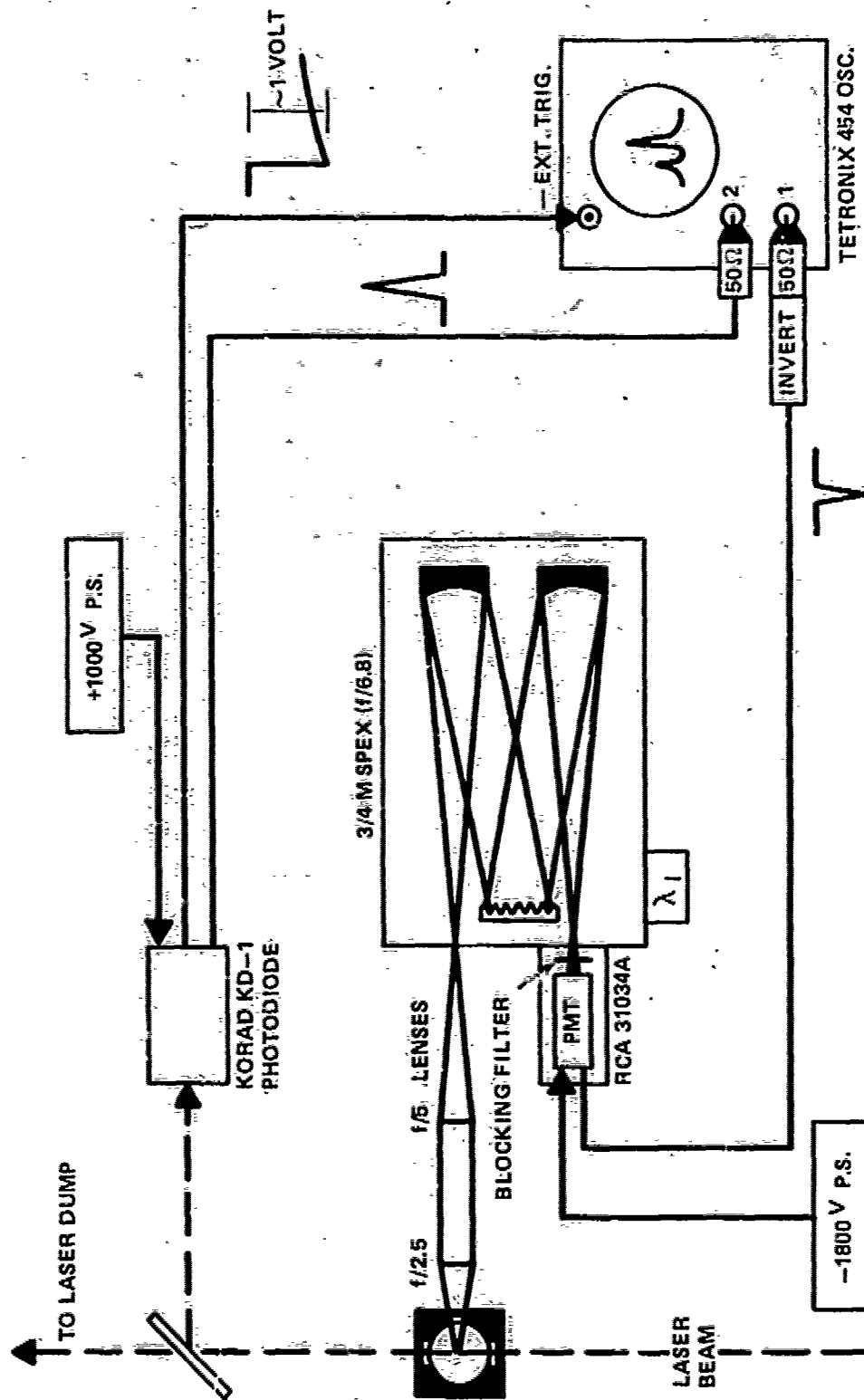


FIG. 8 SCHEMATIC OF ELECTRO-OPTICAL COMPONENTS OF DATA GATHERING SYSTEM

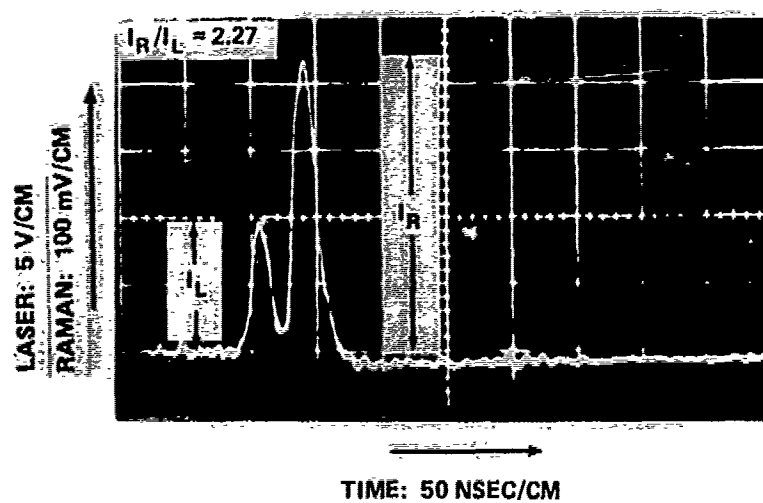


FIG. 9 OSCILLOSCOPE TRACE OF RAMAN AND LASER  
OUTPUT ( $N_2$  IN AIR -  $P_{AIR} = 200$  TORR,  $\lambda_A = 8283\text{\AA}$ )

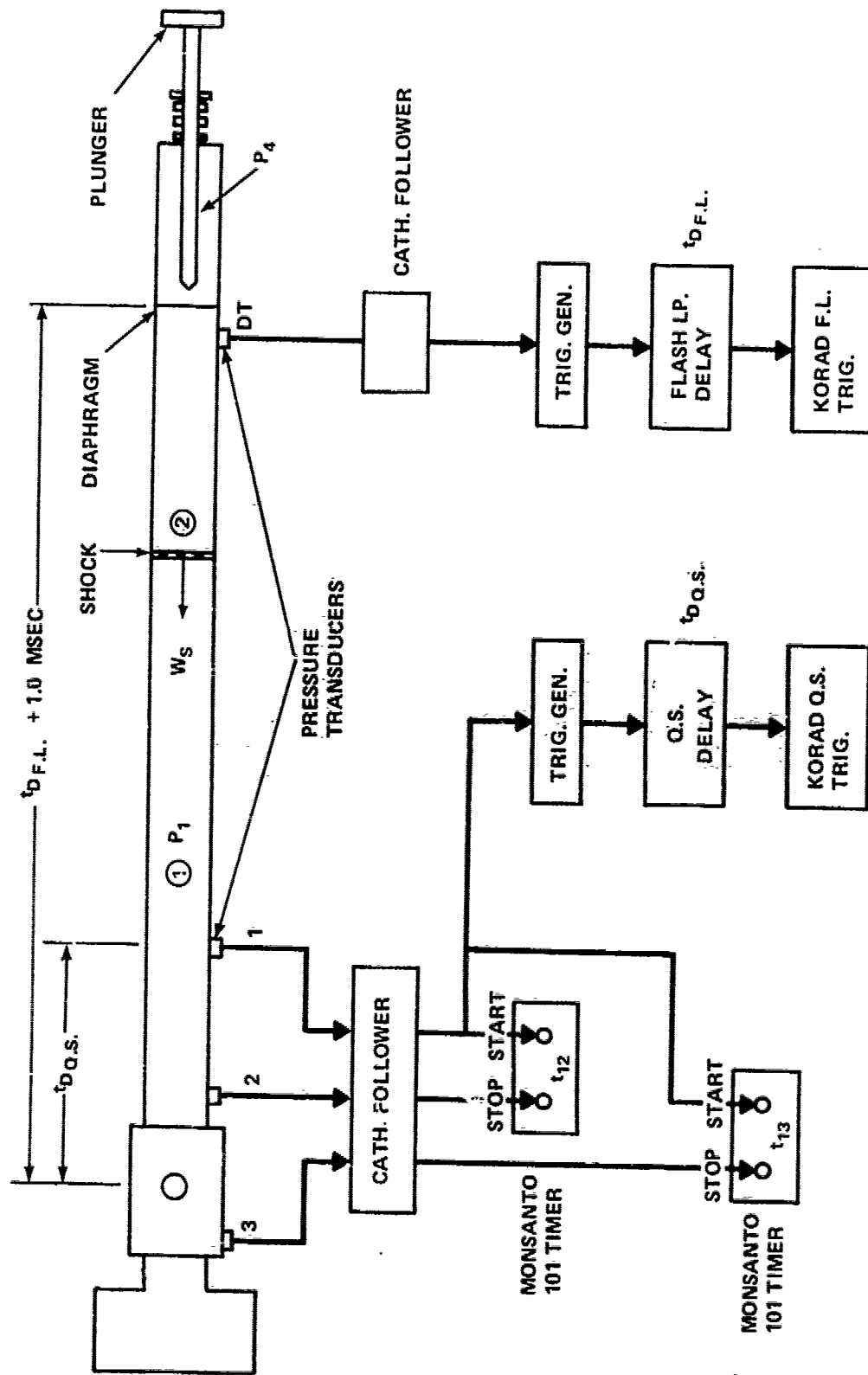


FIG. 10 TIMING CIRCUITRY FOR RAMAN SHOCK-TUBE EXPERIMENT



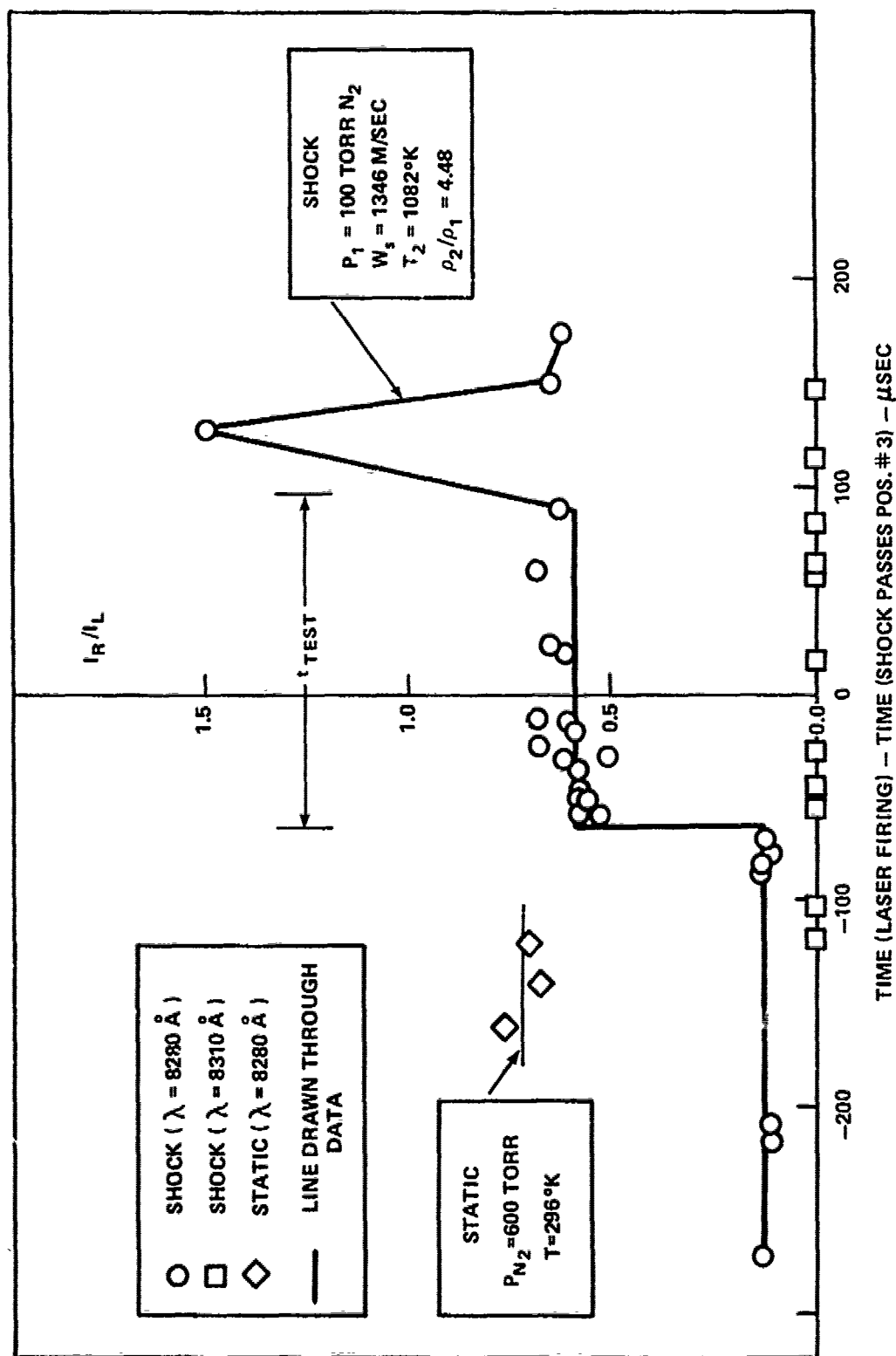


FIG. 11 TIME HISTORY OF RAMAN SCATTERING IN A SHOCK-TUBE EXPERIMENT

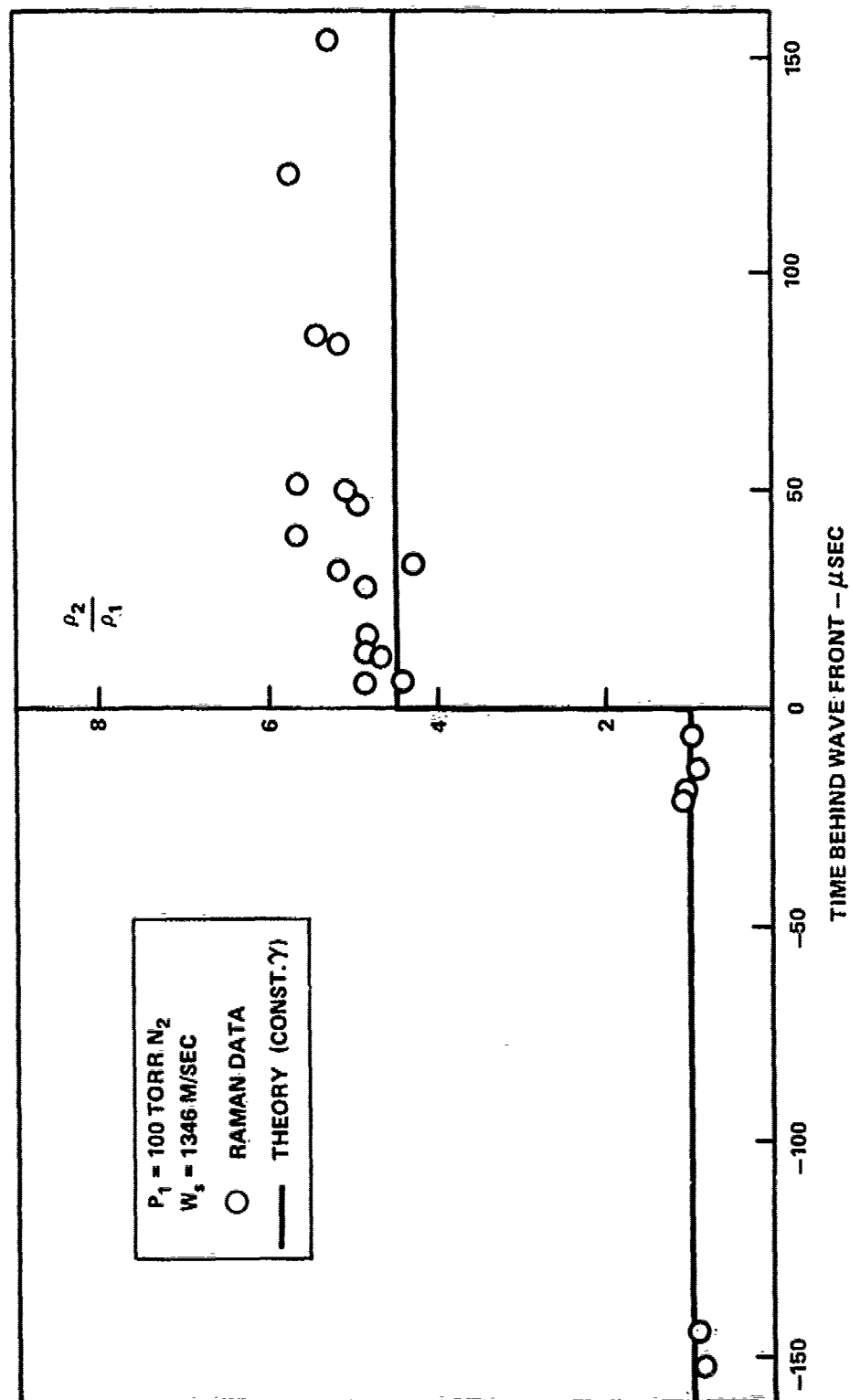


FIG. 12 DENSITY RATIO AS A FUNCTION OF TIME BEHIND THE WAVE FRONT.

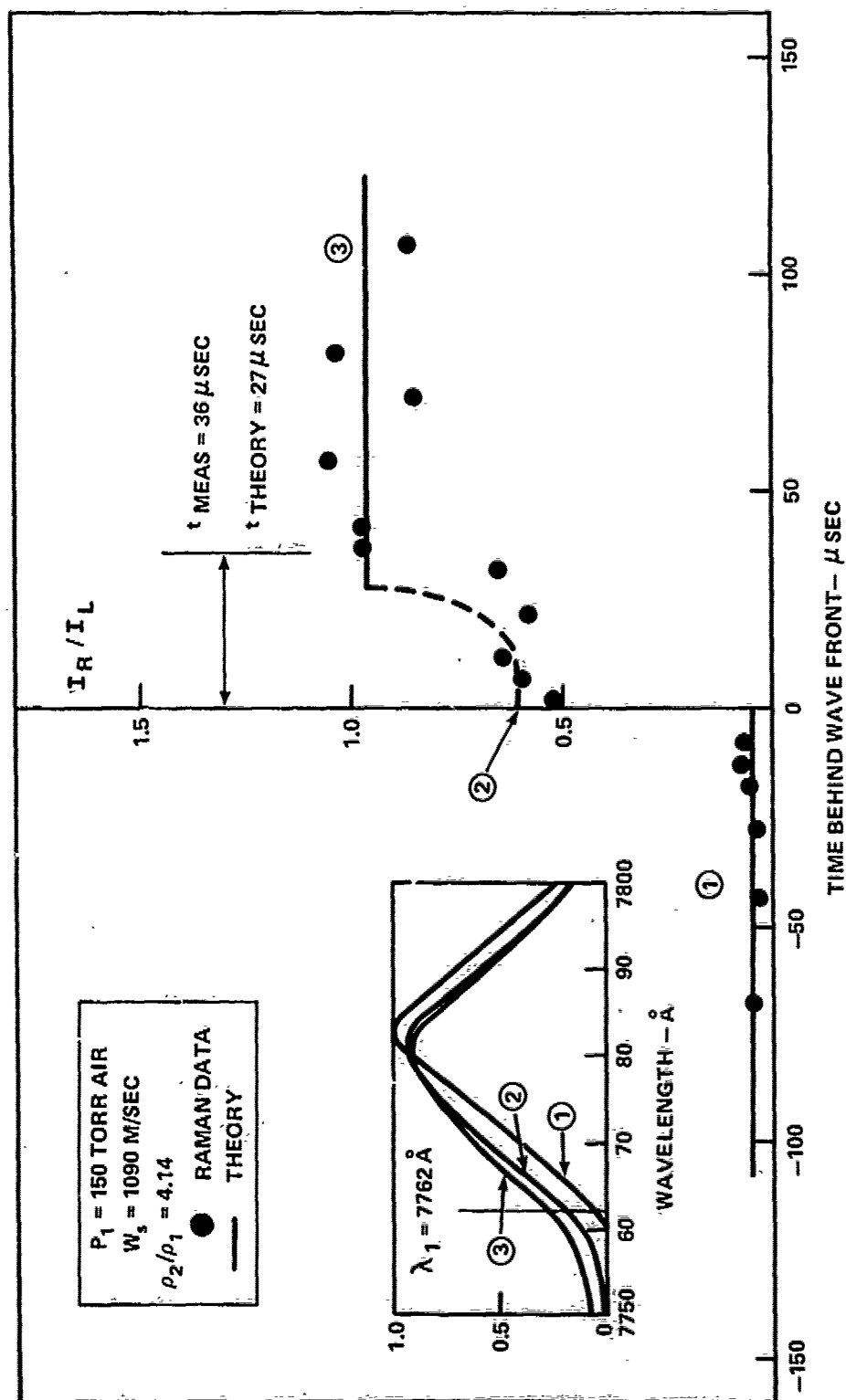


FIG. 13 TIME HISTORY OF RAMAN SCATTERING FROM OXYGEN BEHIND A SHOCK WAVE IN AIR.

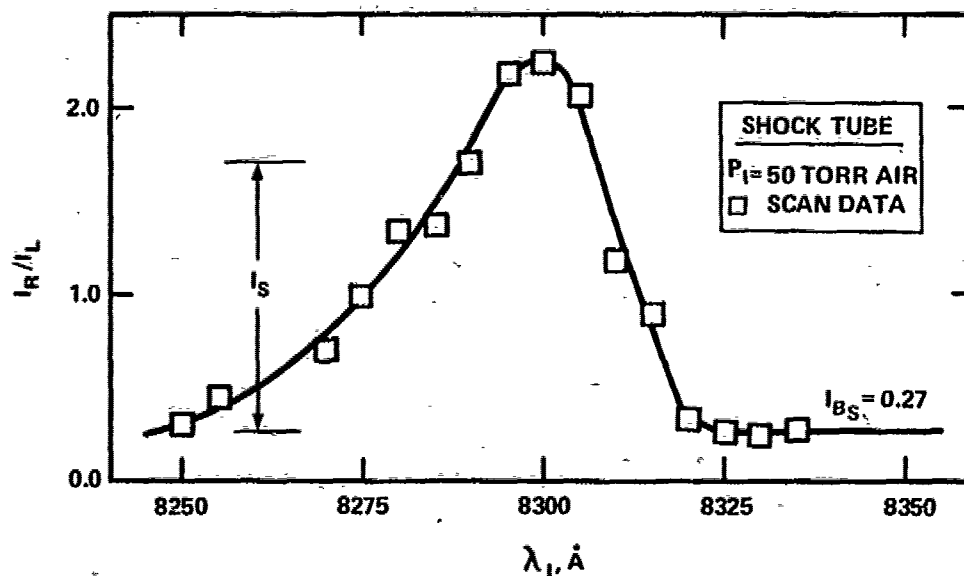
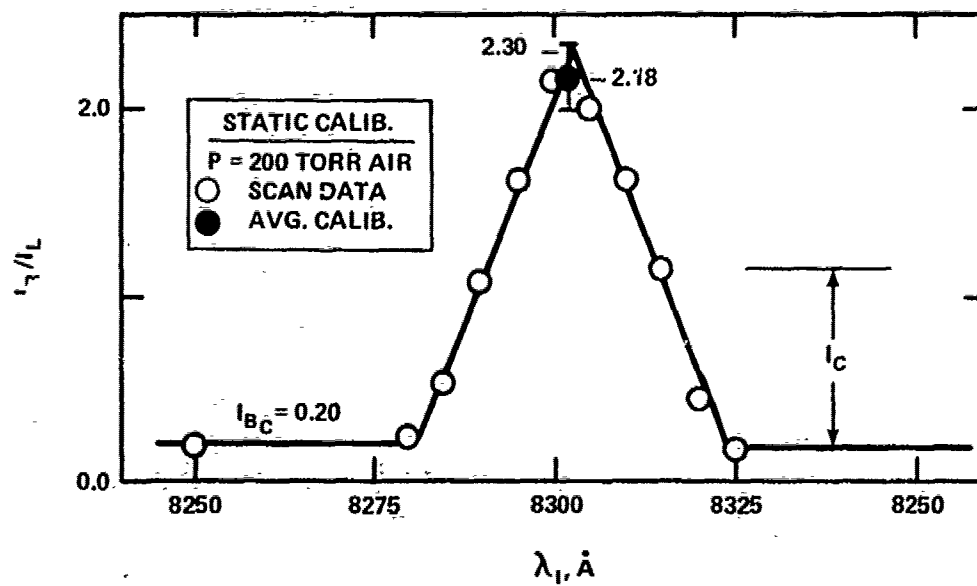


FIG. 14 RAW DATA FOR SCAN METHOD OF DETERMINING DENSITY AND TEMPERATURE USING LRS.

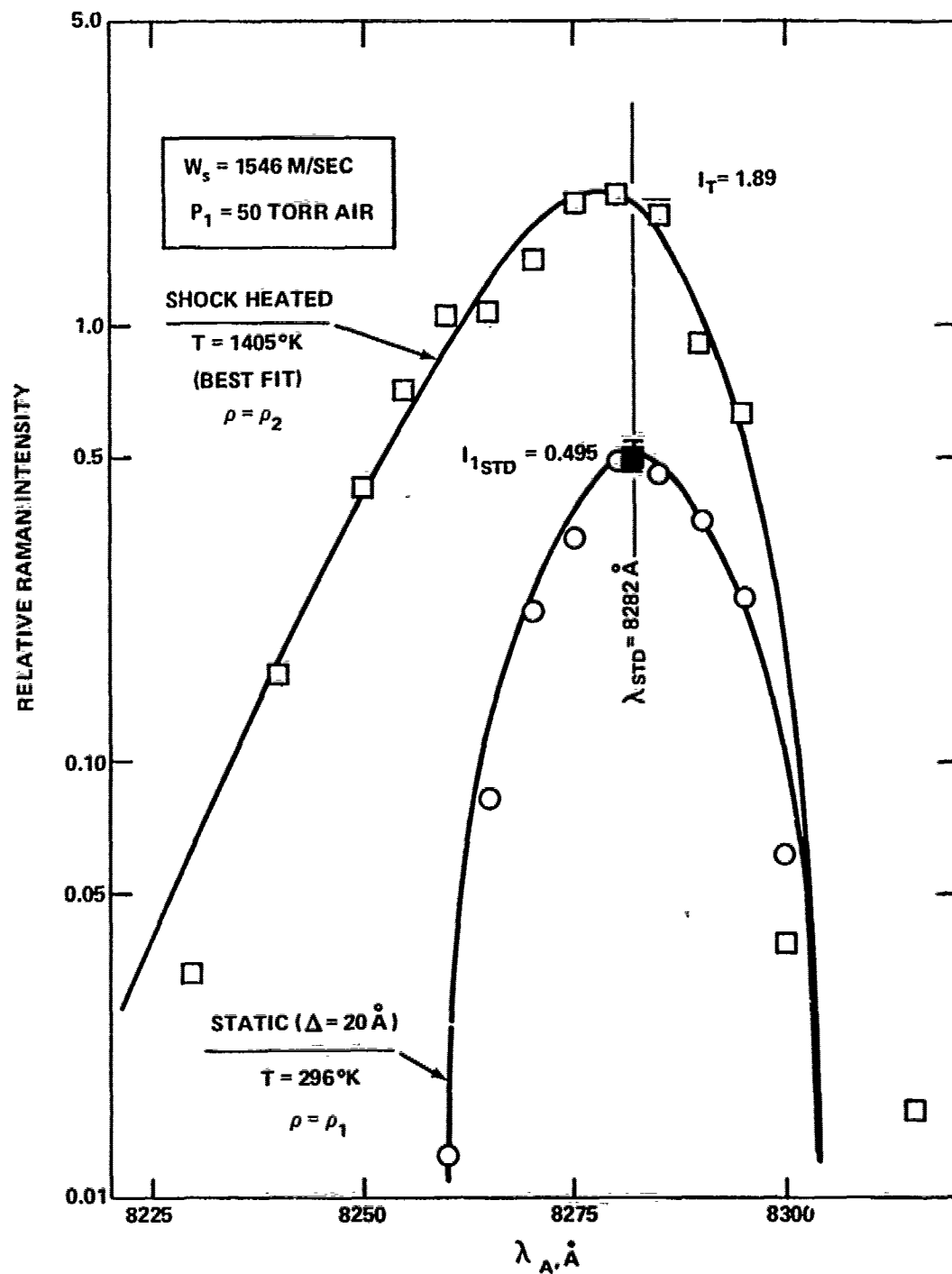


FIG. 15 REDUCED DATA FOR SCAN METHOD OF DETERMINING TEMPERATURE AND DENSITY USING LRS.

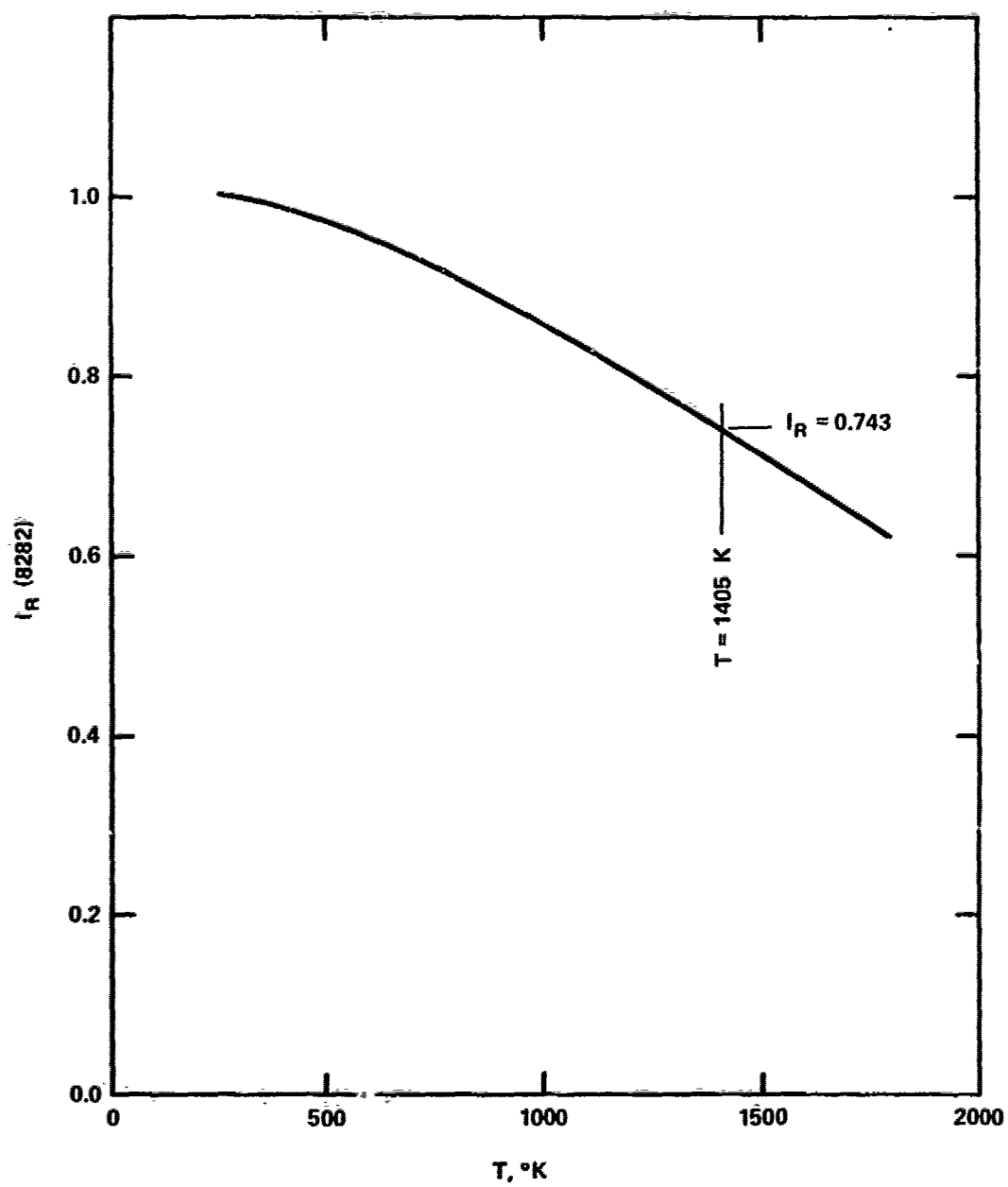


FIG. 16 THEORETICAL CURVE USED TO OBTAIN TEMPERATURE CORRECTION FOR DENSITY DETERMINATION.

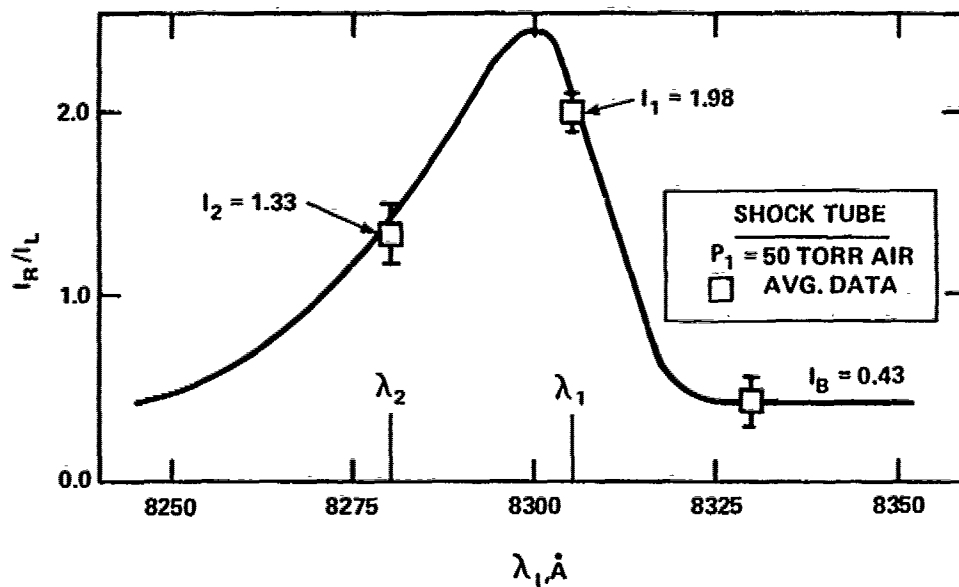
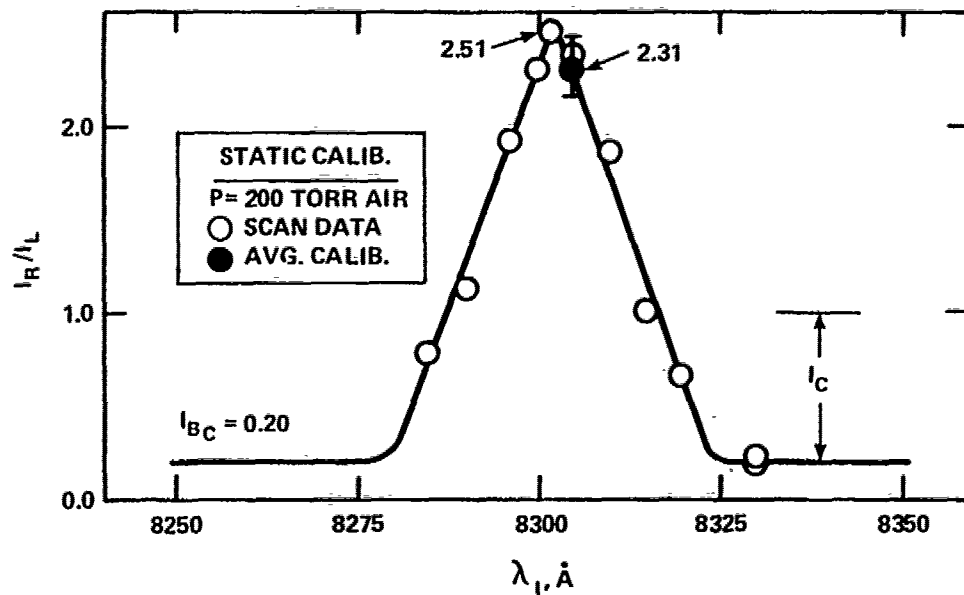


FIG. 17 RAW DATA FOR 2-COLOR METHOD OF DETERMINING DENSITY AND TEMPERATURE USING LRS.

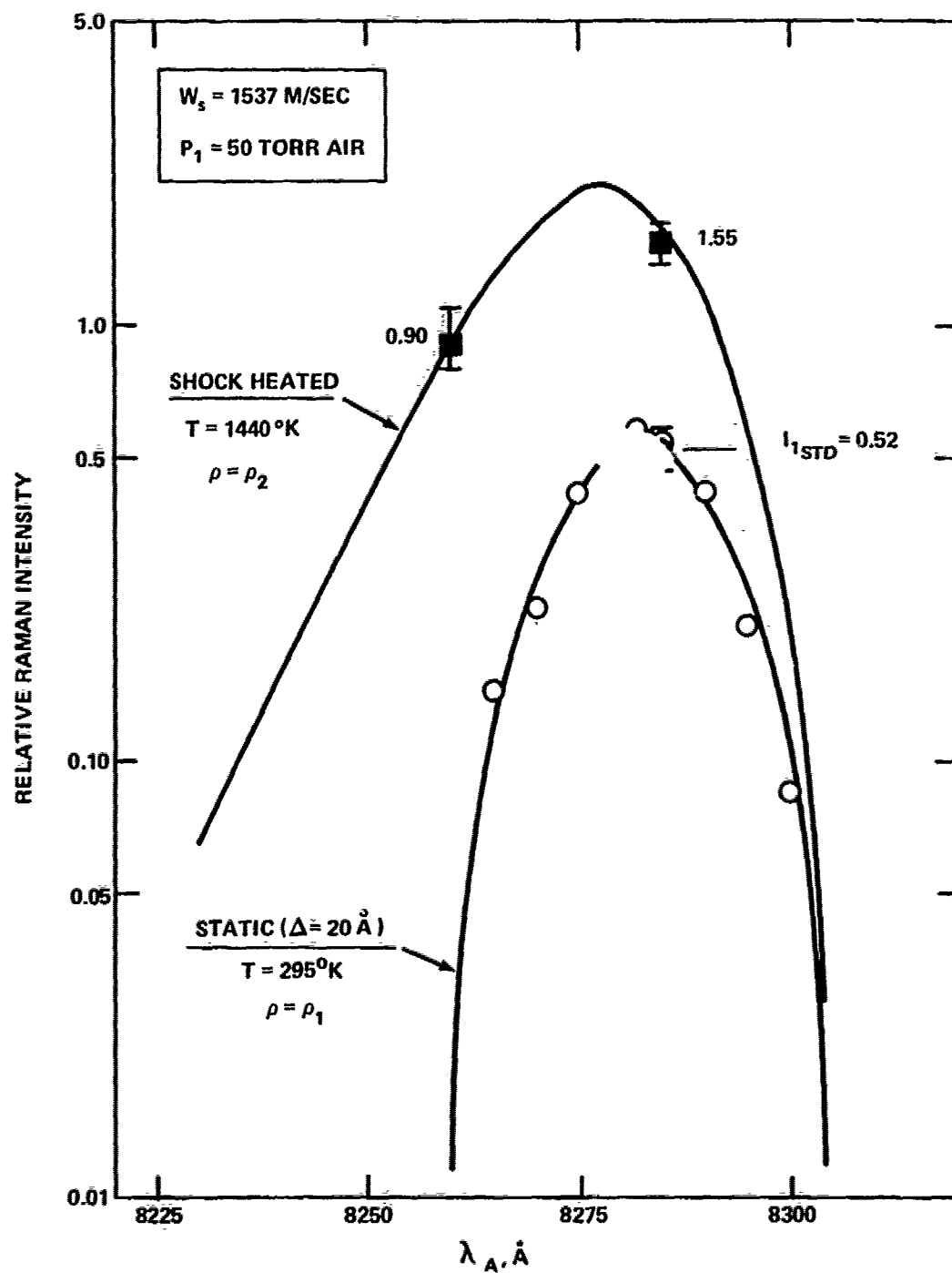


FIG. 18 REDUCED DATA FOR 2-COLOR METHOD OF DETERMINING TEMPERATURE AND DENSITY USING LRS.



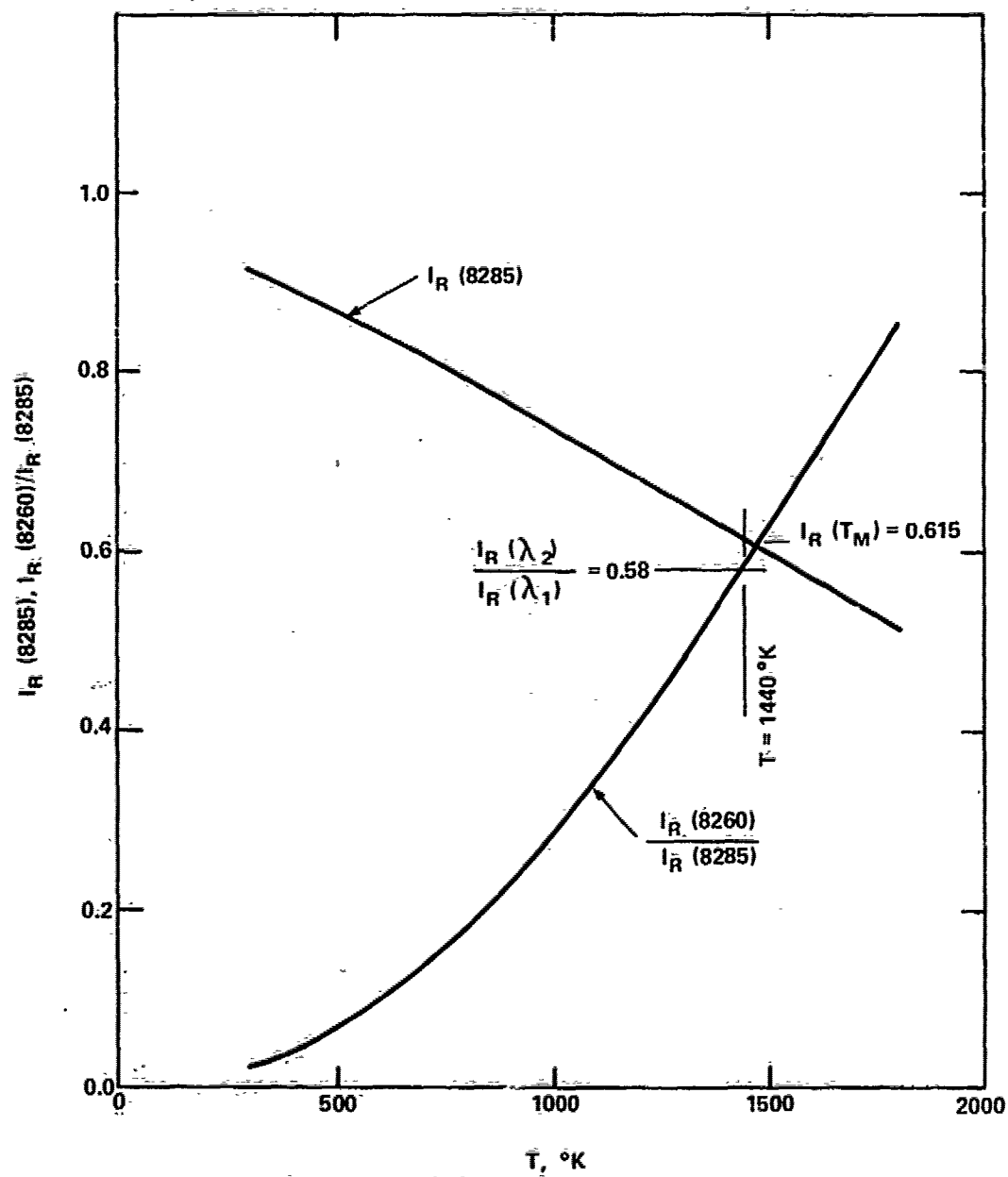


FIG. 19 THEORETICAL CURVES USED TO OBTAIN  $N_2$  TEMPERATURE AND TEMPERATURE CORRECTION FOR DENSITY DETERMINATION

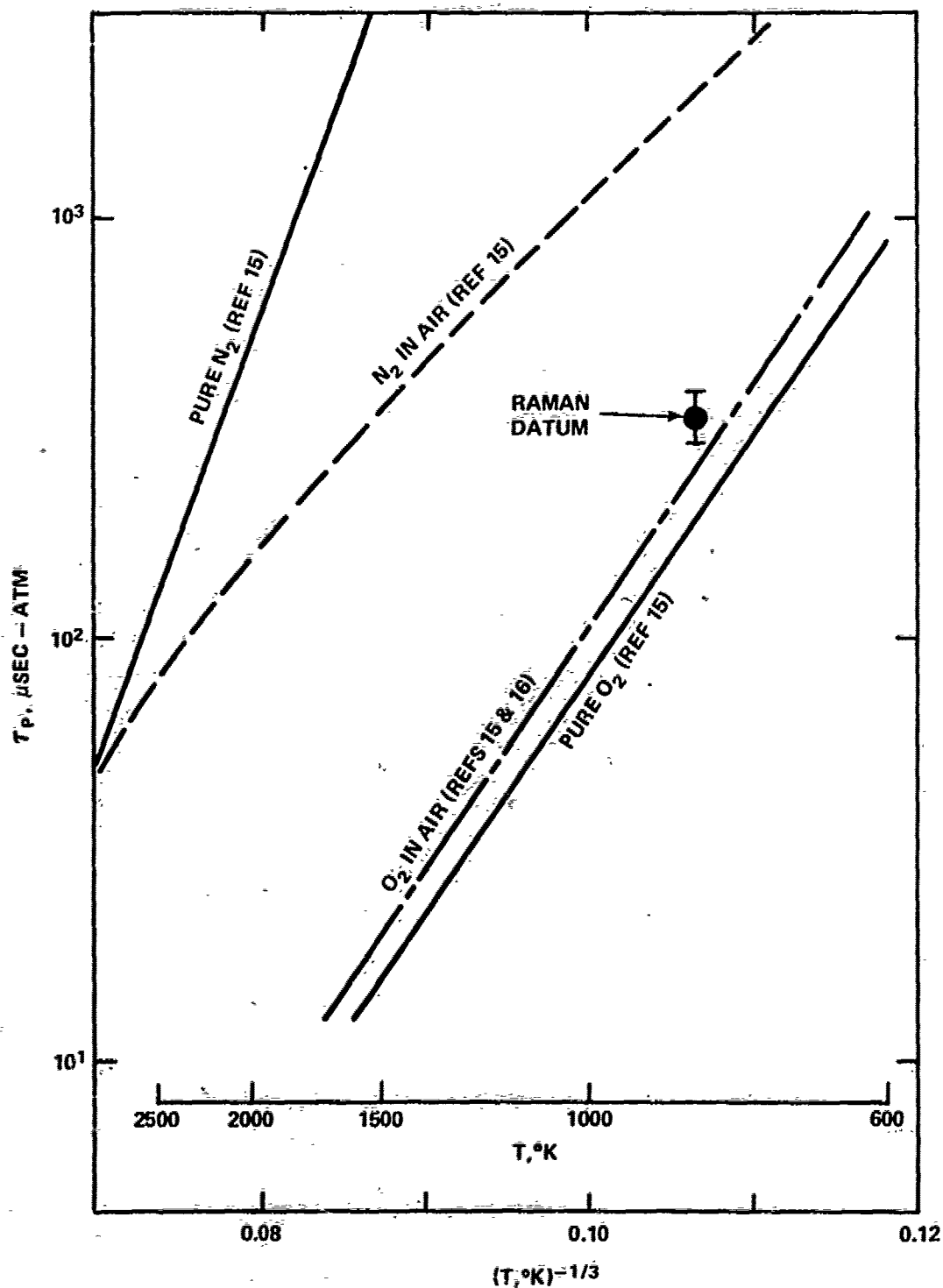


FIG. 20 COMPARISON OF A RAMAN  $\text{O}_2$  IN AIR V-T EXCITATION TIME MEASUREMENT WITH OTHER SHOCK-TUBE DATA.

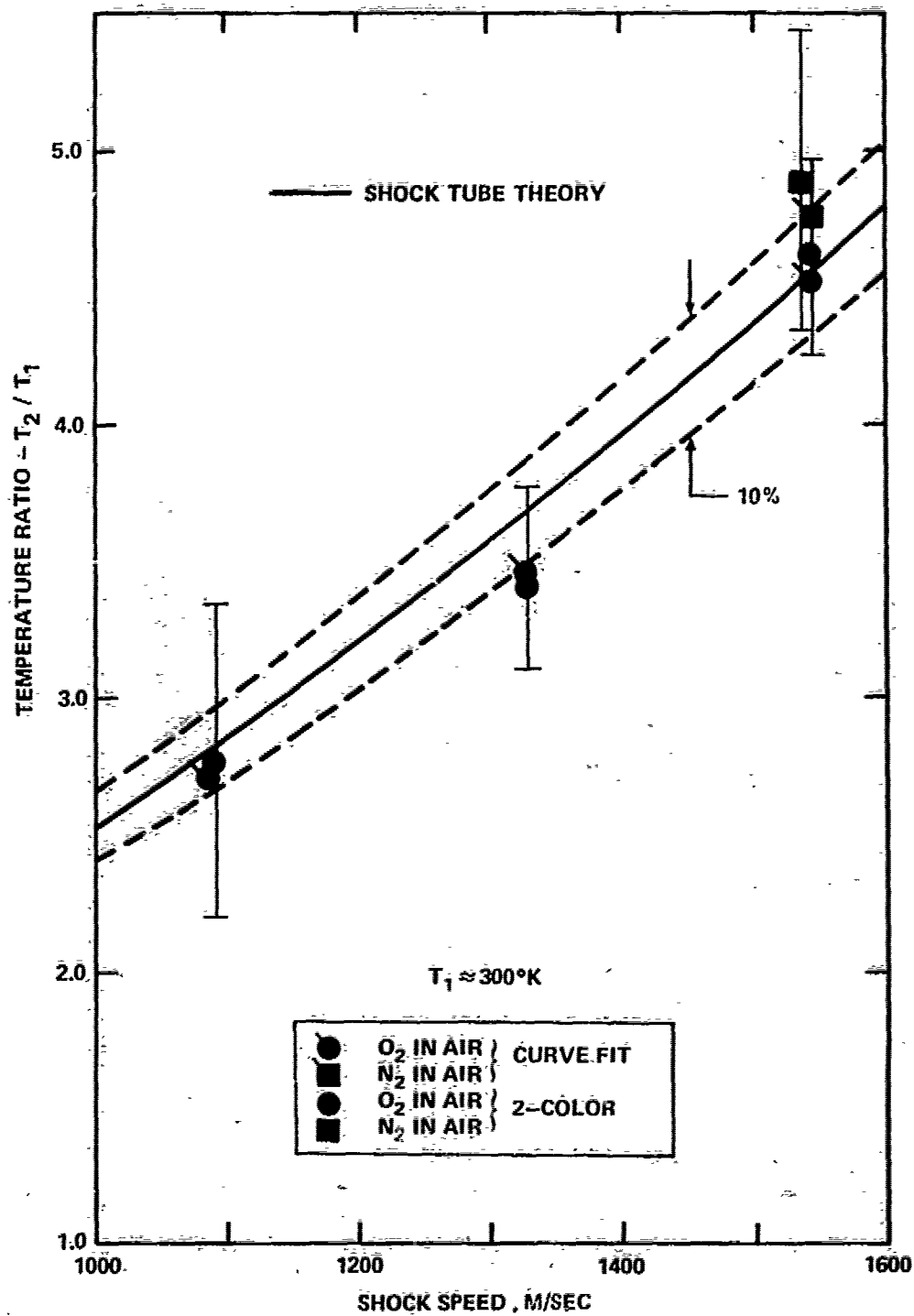


FIG. 21 RAMAN TEMPERATURE RATIO MEASUREMENTS COMPARED TO THEORY FOR  $O_2$  AND  $N_2$  IN SHOCK-HEATED AIR

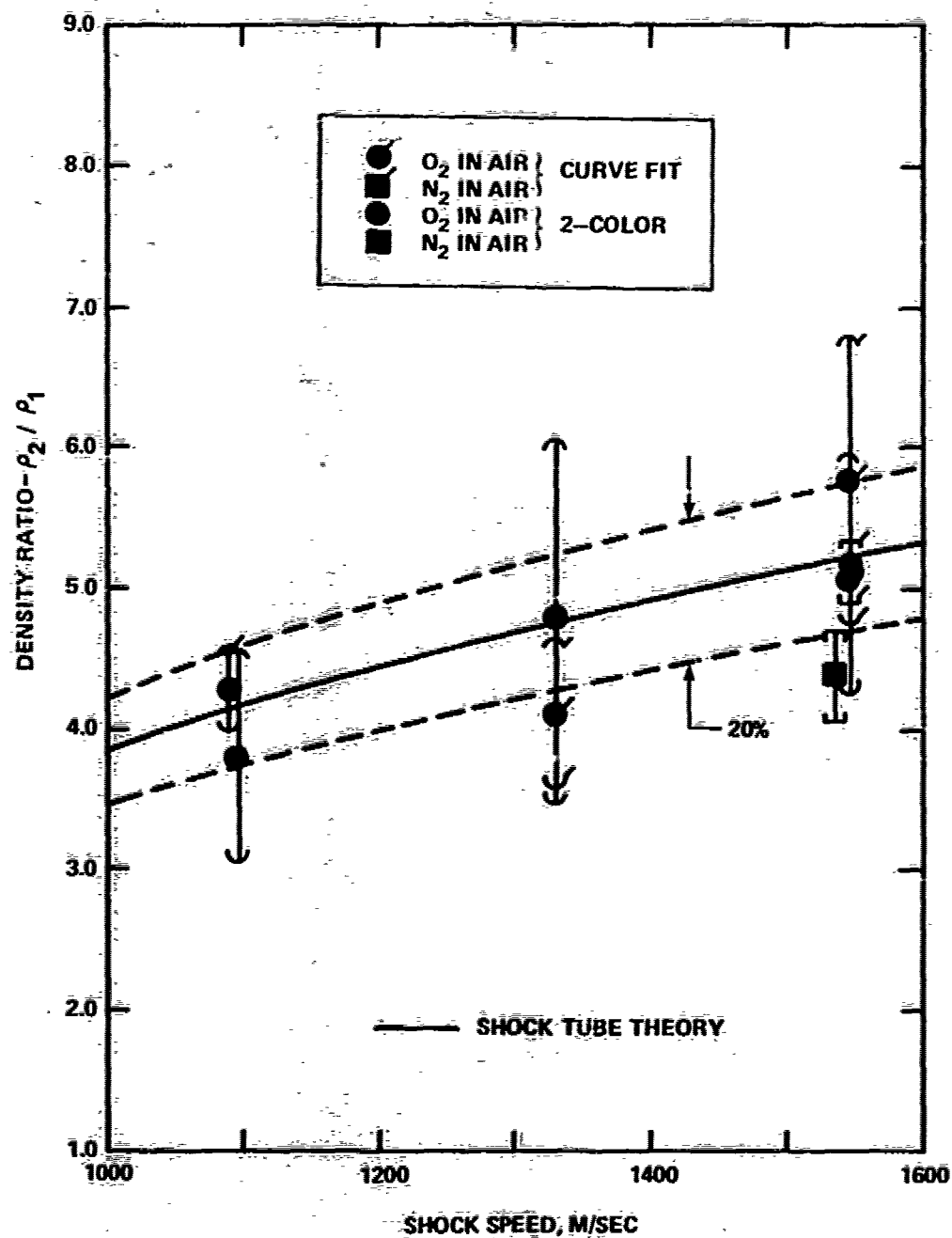


FIG. 22 RAMAN DENSITY RATIO MEASUREMENTS COMPARED TO THEORY FOR O<sub>2</sub> AND N<sub>2</sub> IN SHOCK-HEATED AIR.

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